

HYDROGEOCHEMICAL MODELING OF GROUNDWATER OF THE QUATERNARY AQUIFER IN MELLAGOU VALLEY -BOUHMAMA- (NORTHEASTERN ALGERIA)

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Abstract: The use of groundwater in Bouhmama area for drinking and irrigation purposes has increased progressively over the past decade. The water quality of the aquifer has been significantly altered by anthropogenic activity, such as the extensive use of chemical fertilizers. This research intends to enhance our understanding of the hydrogeochemical function of the Mellagou aquifer in the Bouhmama area in northeastern Algeria. It contributes to the identification of the phenomena affecting the mineralization and the mechanisms of salinization of groundwater. A total of 15 groundwater samples were collected during the rainy seasons in 2022 distributed in the study area. In order to performed the results obtained this study used hydrochemical diagram (Piper, Schoeller, Stiff, Stabler, and Wilcox diagrams), thermodynamic models (saturation indexes), and statistical method (PCA). Three chemical facies with a predominance of the calcic bicarbonate facies reflect the influence of intercalations within the bicarbonate sedimentary formations. Groundwater mineralization is mainly governed by the dissolution and the precipitation of minerals (gypsum and halite, calcite and dolomite), Base Exchange, and anthropogenic activities. The results acquired by PCA show that the variables controlling water mineralization are Cl^- , Na^+ , and K^+ .

Keywords: North-East Algeria, Mellagou Valley, Hydrogeochemical modeling, Principal component analysis, Mineralization.

1. INTRODUCTION

The quality and quantity aspect of groundwater has become more important, as good quality water can be used for various purposes such as drinking water supply, agriculture, industry, and recreation. (Tiri et al., 2014).

Therefore, access to water and the monitoring program that needs to be well-structured are key factors in the economic development of the country (Barbieri et al., 2019; Gaagai et al., 2017).

Problems caused by climate change are likely to be extreme in regions with a semi-arid climate. This is because high levels of evapotranspiration occur in these areas. Drought affects the water cycle, reducing the amount of water available from natural sources (Fletcher et al., 2013; Semadeni-Davies et al., 2008; Chang & Bonnette, 2016; Nistor et al. 2019).

In endorheic watersheds, groundwater is the

most important resource that can be used for drinking and irrigation. In general, groundwater quality is controlled by precipitation, recharge quality, accessibility of surface water, hydrogeochemical processes occurring between water and aquifers, human activities, and unplanned irrigation (Singh & Kumar, 2010; Venkateswaran & VEDIAPPAN, 2013; Nagaraju et al., 2015).

Groundwater contains dissolved salts, which are mainly regulated by the water supply, lithology, and residence time of the water in the aquifer. An excess of dissolved ions in water can affect water quality. Hydrogeochemical studies are particularly useful in assessing, protecting, and managing water resources in terms of water quality identification and assessment (Nagaraju et al., 2015; Balaji et al., 2018; Adimalla et al., 2020).

The degradation of the physicochemical quality of water resources in aquifers of semi-arid

zones is the subject of future challenges. Climate variability and change affect the recharge and discharge of aquifers and, consequently, their overall groundwater reserves (Lăcătușu, et al., 2019; Jeff Garçon et al., 2020).

Salinity is considered one of the main causes of groundwater quality degradation worldwide, (Kloppmann et al., 2011).

The water needs of the Bouhmama region are provided by groundwater from the Plio-Quaternary aquifer. The water of the aquifer is accessible through boreholes. This aquifer has been excessively exploited in recent years to support the development of irrigated agriculture, and the needs of the population of the region. This defines a state of depletion of reserves as the traditional wells. This increase has caused pressure on groundwater resources, also related to climate, the feeding of the water table by surface water leads to a change in the original chemical composition. This condition, associated with drought, has caused the drying up of many springs and induced degradation of chemical quality, illustrated by the increase in salinity of the waters (Khemmoudj et al., 2016; Barbieri et al., 2019; Fathi et al., 2017; Bencer, 2005; Zubair et al., 2022).

Tracing and determining the origin of variation in water chemistry is often related to natural causes (climate change, erosion, and weathering of crossed formations) and anthropogenic activities (urban, industrial, and agricultural) (Csete et al., 2021; Kumar et al., 2019).

According to (Gomo et al., 2013), some hydrogeochemical processes occur during this evolution, which should be identified.

This hydrogeochemical study aims to identify the processes responsible for the evolution of groundwater chemistry in the aquifer of Mellagou

Valley, and the mechanisms of salt load acquisition.

2. MATERIAL AND METHODS

2.1. Study area

The study area is located in the northeastern part of the Algerian Territory (Figure 1), in the center of the mountainous region of the Aures. It is part of wadi Mellagou sub-watershed, which covers an area of 577 Km² (Figure 1). Dominated in the North - West by the mountainous massif of Beni Imloul - Chelia which culminates in the highest peak of Northern Algeria "Ras Kelthoum", With an altitude of 2326 m, and the massif of Ouled Yagoub - Beni Oudjana in the North - East of the sub-catchment which culminates at 2143 m in Bazaz Mountain (Lessard, 1952; Houbib, 2013). The climate of the study area is semi-arid, characterized by the alternation of a hot, dry season and a cold, wet season (Gaagai et al., 2022).

According to the geological map of the Mellagou watershed (Figure 2). The sub-basin is formed mainly by permeable to semi-permeable formations that occupy almost the entire area of the basin, which promotes the infiltration of surface water.

The study area is characterized by a stratigraphic series extending from the Triassic period consisting of clay, gypsum, dolomite, and limestone to the Quaternary period formed mainly by limestone and marl (Lafitte, 1939; Lessard, 1952).

Two major aquifers are highlighted:

A shallow aquifer occurring in Quaternary alluvial formations underlain by clays and marls that are sometimes gypsum; and a deep Cretaceous aquifer in fractured limestones that outcrop on the boundaries and have been recognized with deep drilling (Houha et al., 2016).

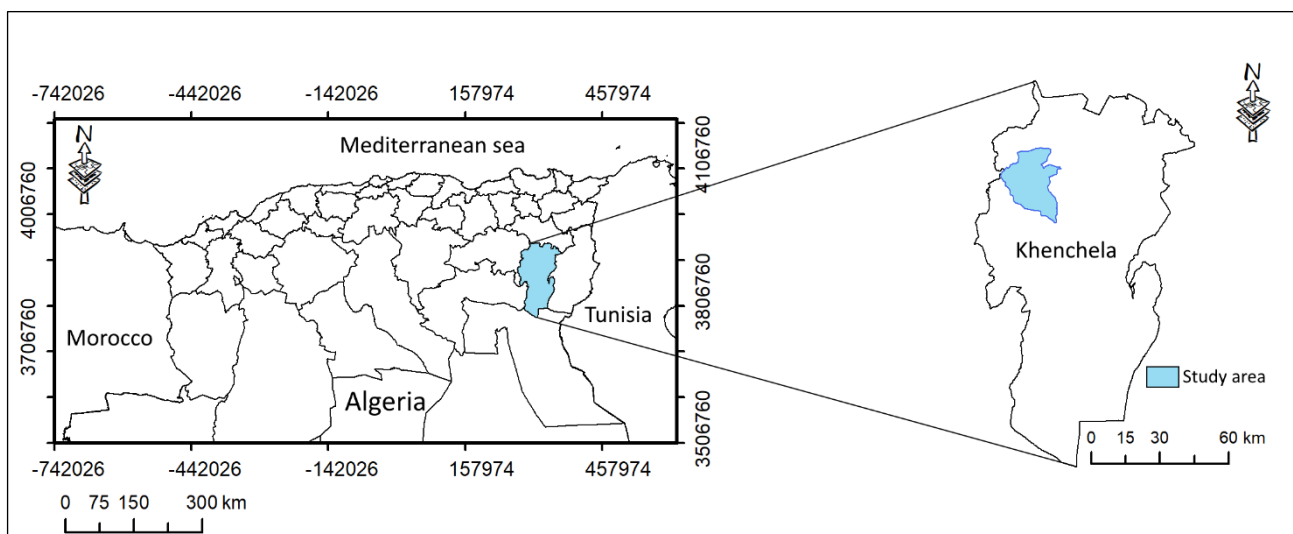


Figure 1. The geographical location of the study area

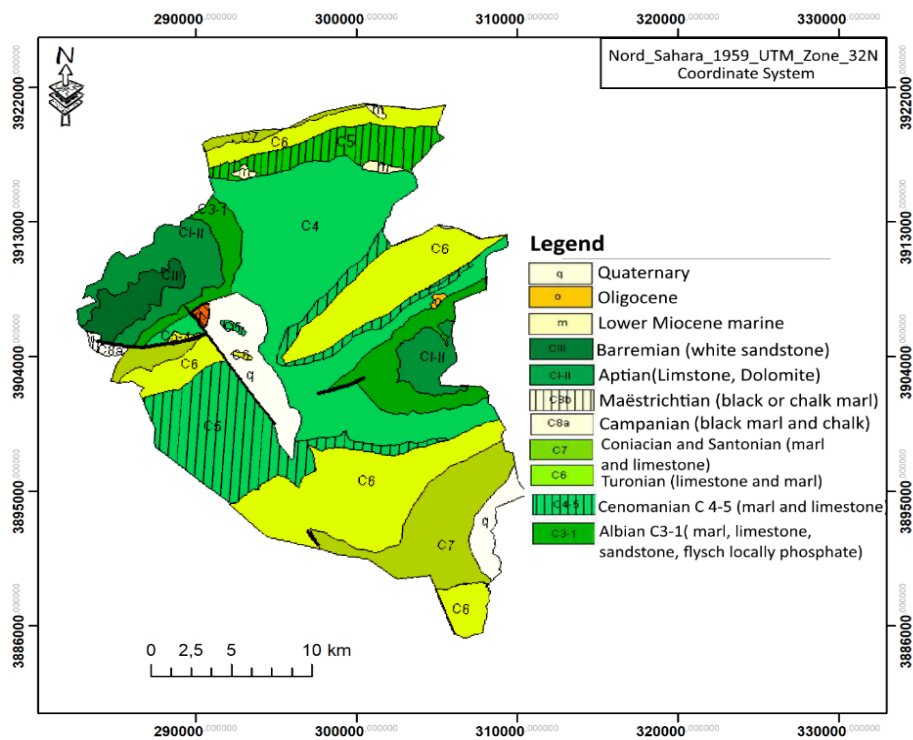


Figure 2. Geological map of the study area (Laffite, 1939; in Houbib, 2013; modified)

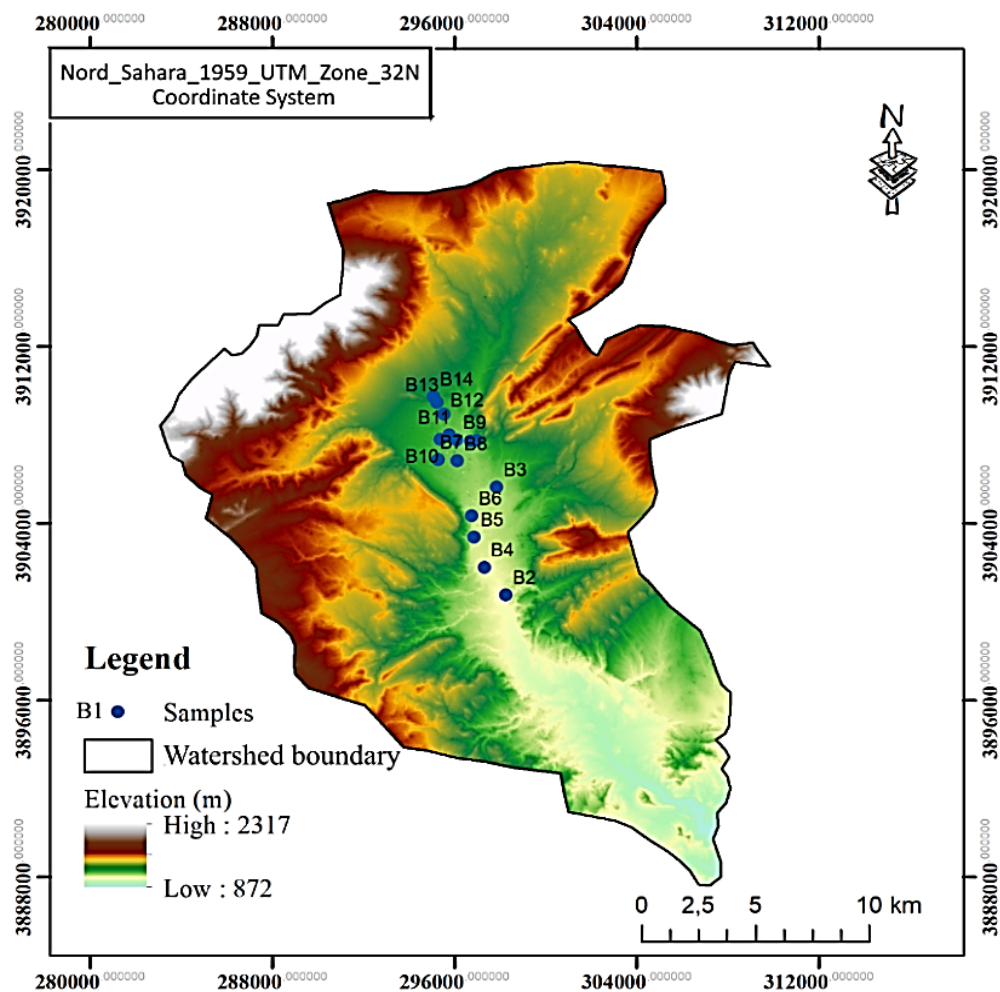


Figure 3. Inventory map of the study area, May 2022

2.2. Sampling and analytical methods

Seasonal monitoring of the physicochemical parameters of the groundwater of the Quaternary aquifer was carried out during the wet period of the year 2022. The water sampling concerned 15 boreholes, well distributed in the aquifer (Figure 3).

The water points were located by their geographical coordinates determined by a GPS-type Garming. The physical parameters: temperature ($T^{\circ}\text{C}$), pH, electrical conductivity (EC), and TDS, were measured in situ using a multi-parameter brand YSI (Pro DSS).

The chemical analysis of major ions in the water was carried out at the laboratory of the Algerian Water Company. The HCO_3^- , Ca^{2+} , Mg^{2+} , and Cl^- ions were measured by the titration method, while the other ions NO_3^- , SO_4^{2-} , K^+ and Na^+ were measured using a UV spectrophotometer.

The realization of the maps was done with the ARC GIS 10.8 software (ESRI).

The thermodynamic modeling was done with the software DIAGRAMME, version 6.77

The calculation of the saturation indexes SI was done with the software PHREEQC 3.7

The descriptive statistics were done with XLSTAT 2015 software.

3. RESULTS AND DISCUSSION

3.1. Piper and Stiff diagrams (chemical facies)

The analysis of the Piper diagram has allowed us to highlight the different chemical facies of the waters, (Piper, 1944). The analysis of chemical parameters shows that the waters of the Plio-Quaternary aquifer are characterized by the abundance of ions (HCO_3^- , Ca^{2+} , Na^+ , Cl^-).

Plotting the major ions of groundwater on the Piper diagram for the wet period of the year 2022, (Figure 4) has identified three main types of chemical facies:

-Sodium chloride waters. According to (Debièche, 2002): The existence of these two elements Na and Cl are related to the dissolution of evaporite formations rich in halite.

-Calcium sulfate waters and calcium bicarbonate waters are represented by the majority of the water points, (Figure 4a).

Stiff diagram allows us to obtain a polygonal geometric (Figure 4b) characteristic of a given water family. Major ion analysis data are plotted on horizontal axes on either side of the original vertical axis. The cations are shown on the left ($\text{Na}^+ + \text{K}^+$,

Ca^{2+} , and Mg^{2+}), and anions on the right (Cl^- , HCO_3^- , SO_4^{2-} , NO_3^-) (Stiff, 1951).

The interpretation of Stiff's diagram obtained from the water samples (Figure 4b) allowed to divide of the groundwater according to its mineralization into 2 groups presenting:

- Medium mineralization: deep bicarbonate sandstone boreholes

- Low mineralization: superficial boreholes.

This differentiation of mineralization has been studied by Ligban et al., 2009. It could reflect a more prolonged residence time for water from deeper boreholes compared to water from more superficial boreholes.

3.2. Schoeller diagram

In Schoeller's representation (Figure 4c), it is noticeable that the straight lines intersect each other, which translates that the waters are of different origins; even if there are waters that have the same chemical facies (Schoeller, 1977).

3.3. Stabler diagram

In the light of figure d4. The Stabler diagram is established from the HCO_3^- ions.

We notice that the contents of bicarbonates, calcium, and chloride, are high but those of sulfates, magnesium, sodium, are the least. It can be seen that the alkalinity level is also high.

3.4. Wilcox diagram

Wilcox in 1948 established this diagram used to assess the risk of salinization of soils. It uses the electrical conductivity (EC) or total dissolved charge (TDS).

According to Wilcox (1948), waters are classified from their proportion of sodium according to the electrical conductivity. We note that the groundwater of the aquifer has a sodium content that varies between 27 and 70%. Therefore, it is noted that these waters as well as the soil of the catchment area of the study area present a risk of salinization. (Figure 4e).

3.5. Origin of the mineralization of the groundwater of the Mellagou valley

3.5.1. Binary relationship

We have represented the major elements as a function of chloride to understand the process of mineralization of groundwater, (Figure 5). The chloride is a conserved element, does not anticipate water-rock interactions, characterizes the origin of the

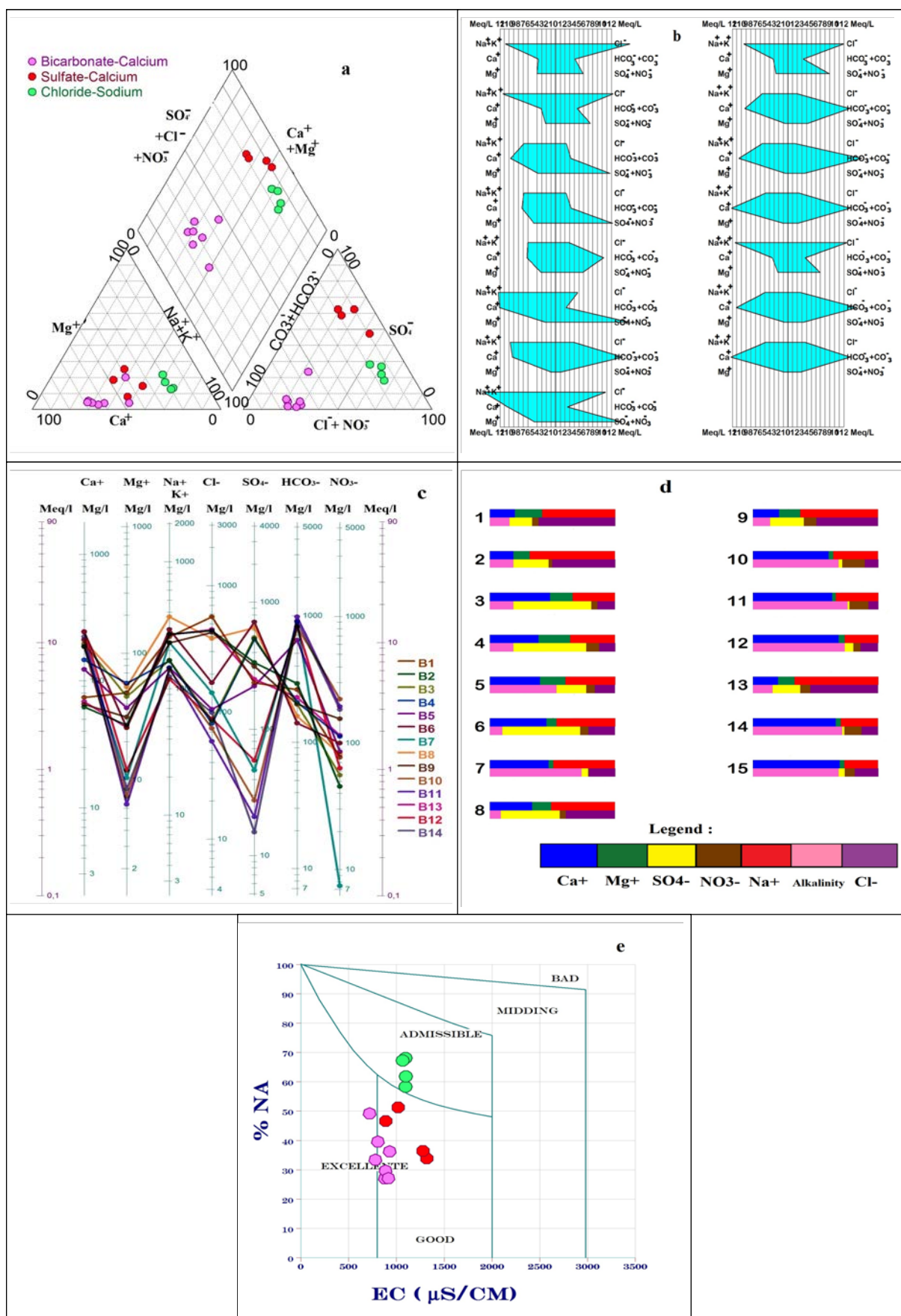


Figure 4. Piper (a), Stiff (b), Schoeller (c), Stabler (d), & Wilcox (e) diagrams of the groundwater of Mellagou Valley, May 2022.

salinity of the water and is a tracer of mixing (Fidelibus & Tulipano, 1996; Azaza et al, 2011).

The Na^+ vs Cl^- plot shows that the majority of points are below the mixing line. Since the Na^+

content should balance the Cl^- content. The Na^+ deficit is explained by the phenomenon of ionic base exchange between water and aquifer and resulting in Na^+ adsorption and Ca^{2+} release. (Figure 5a). These

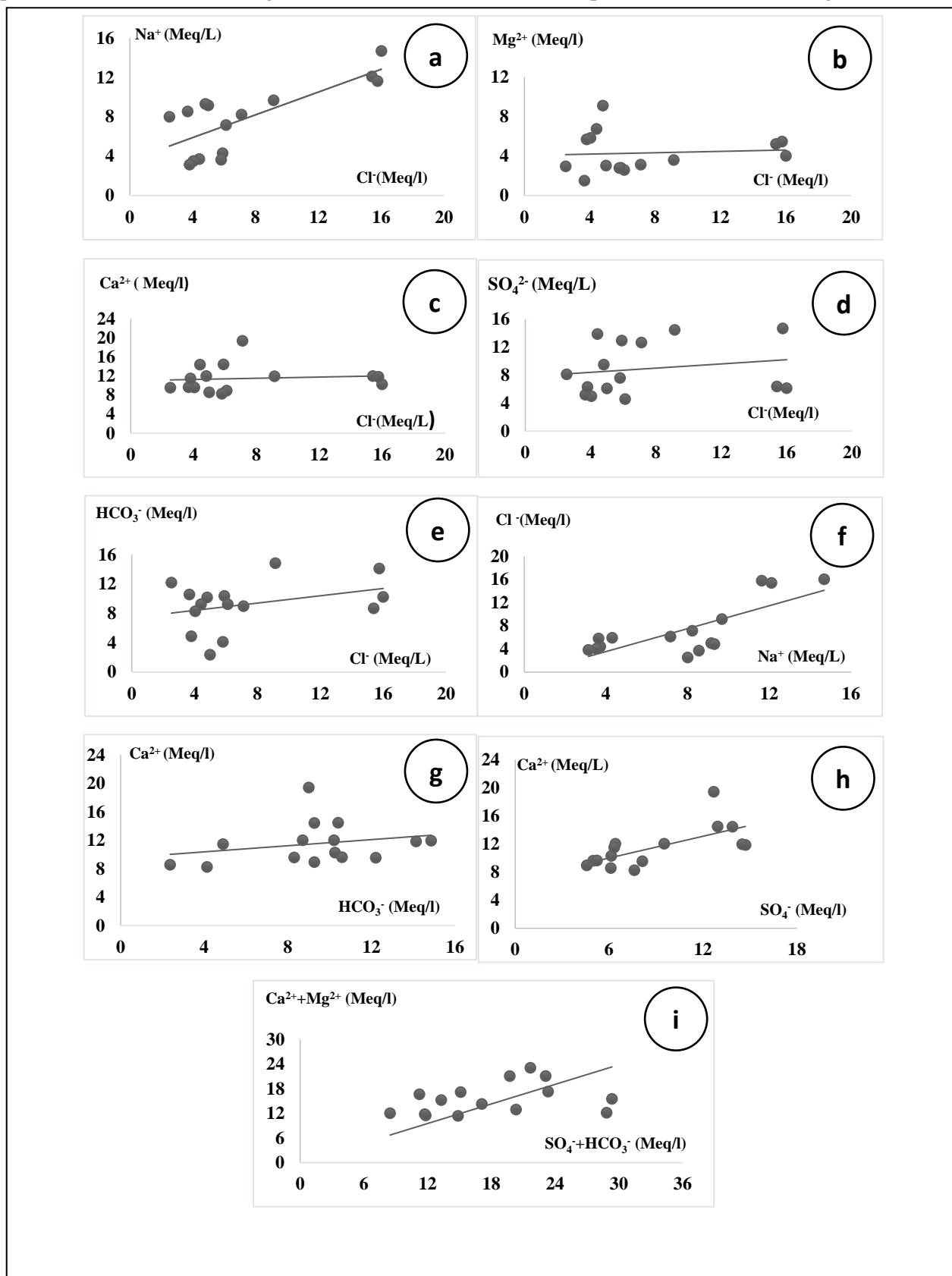


Figure 5. Relationship between major elements of the groundwater of the Mellagou Valley, May 2022.

waters are controlled mainly by cation exchange reactions between water and red Pontian clays resulting in sodium fixation and calcium release (Houha, 2007; Gouaidia et al., 2012; Djenba et al., 2015).

The Ca^{2+} vs. Cl^- (Figure 5c) and Mg^{2+} vs. Cl^- (Figure 5b) diagrams show that the points are often below the mixing line. This observed Cl enrichment is approximated by anthropogenic activities. (Agriculture and household waste), (Drouiche et al., 2022).

The SO_4^{2-} vs Cl^- ratio (Figure 5d) shows that the $\text{SO}_4^{2-}/\text{Cl}^-$ ratio is in favor of sulfates. It can be suggested that this increase in the ratio is due to the leaching of the evaporate layers of the geological bedrock, not to mention the pollution by the return of irrigation water loaded with fertilizers (Drouiche et al., 2022; Panpan et al., 2019).

The HCO_3^- vs. Cl^- ratio (Figure 5e) shows a heterogeneous distribution of bicarbonates due to pH variation and gas exchange with the atmosphere. Bicarbonate excess is evidenced by the dissolution of bicarbonate formations (Zahi et al., 2021; Bouziane & Labadi, 2009).

The Cl^- vs. Na^+ correlation shows a first group, with points below the line, influenced by halite dissolution and a second group has a sodium deficit (Figure 5f). The Na^+ deficit is more or less balanced by a Ca^{2+} enrichment. This trend suggests base exchange reactions by the clay matrix of the surficial aquifer, which may act as a cation exchanger (Capaccioni et al., 2005; Stigter et al., 2006).

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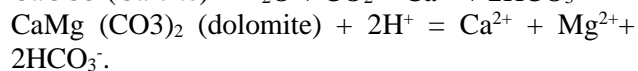
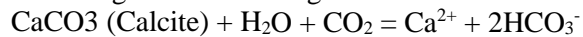
The Ca^{2+} vs HCO_3^- ratio (Figure 5g) shows that the majority of water points fall below the line and the Ca/HCO_3 ratio is in favor of bicarbonates. It can be suggested that this increase in the ratio is due to leaching from carbonate formations (Berkani et al., 2016).

The diagram (Ca^{2+} vs SO_4^{2-}), (Figure 5h); shows two groups of points, a first group where the points align around the slope line influenced by the

dissolution of gypsum and/or anhydrite. A second group formed by more mineralized water points that show an excess of Ca^{2+} which could be due to a process of dissolution of the limestone formations and Base Exchange reactions (Djenba et al., 2015; Zahi et al., 2021).

The examination of Figure 5i shows that the majority of points above the equilibrium line is explained on the one hand by the presence of other evaporites of alkaline origin the gypsum that has dissolved in water. On the other hand, exchange of base is due to an excess of Ca^{2+} and Mg^{2+} coming from the carbonate formations. This explains why the water is in equilibrium mainly with dolomite and aragonite (Drouiche et al., 2022; Gouaidia et al., 2012).

Garrels & Mackenzie (1971) have studied the dissolution of occurs under natural conditions according to the following reactions:



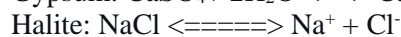
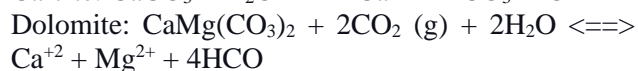
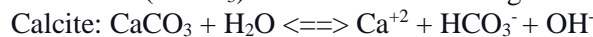
3.5.2. Geochemical modeling

A simple model of the PHREEQC geochemistry software is used to determine the saturation indices of the minerals that drive the mineralization of the Mellagou Plateau groundwater. The results are presented in Figure 6.

An SI value greater than zero indicates that the groundwater is supersaturated with mineral salts and is therefore unable to dissolve many minerals.

Such an index value reflects groundwater discharge from an aquifer containing a sufficient amount of mineral material with enough residence time to reach equilibrium. Nevertheless, supersaturation can also be produced by other factors that include dissolution, common ion effect, evaporation, rapid increase in temperature and dissolved CO_2 concentration.

According to (Ahoussi et al., 2012): the presence of calcium and hydrogen carbonate ions in water is thought to be from the weathering of carbonates (CaCO_3) based on the following formula:



This reaction explains the dominance of the calcium bicarbonate facies in the waters of the basin. This denotes the existence of a chemical differentiation between the sampled aquifers.

For all samples, there is undersaturation of gypsum. The dissolution of gypsum is confirmed by the

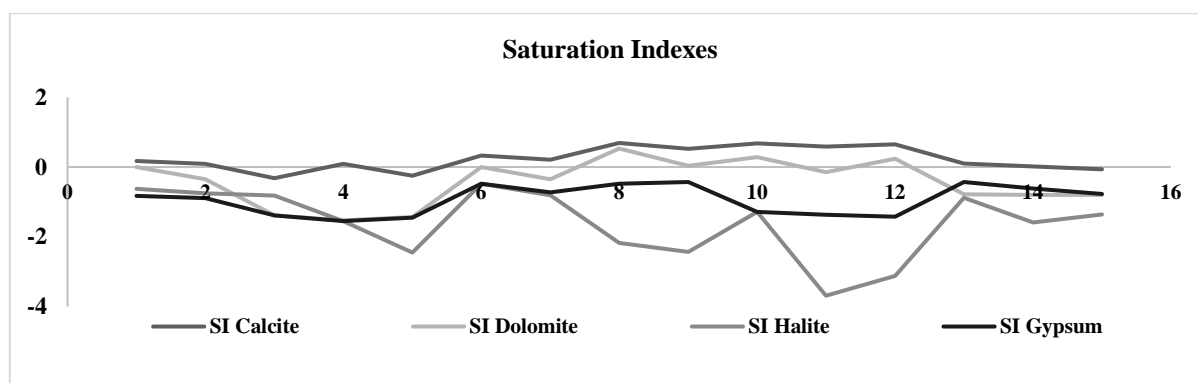


Figure 6. Saturation indexes of the groundwater of the Mellagou Valley, May 2022

inversely proportional relationship in an undersaturated state. The hypothesis of gypsum hydrolysis could be suggested for this purpose in the Quaternary aquifer.

The calcite saturation indices of the waters (Figure 6) concerning the minerals concerned show a state of supersaturation for most of the samples, which confirms the dissolution of the carbonate minerals, with the precipitation of the calcite.

The dolomite saturation indices of most of the samples are positive, which also confirms the hypothesis of the dissolution of these minerals in the water, with the precipitation of dolomite.

The indices of saturation of Halite present a state of under-saturation, which confirms the hypothesis of the dissolution of evaporates (Adimalla et al., 2020; Sedrati et al., 2017; Zahi, 2014).

On the other hand, the leaching of salts by rainy events and the return of irrigation waters favor the accumulation of salts in the soils and contribute to the acquisition of mineralization and the increase of the saline load of the waters of this aquifer.

3.5.3. Statistical analysis

The physicochemical parameters of the borehole waters were subjected to a principal component analysis (PCA) to determine the different processes involved in the mineralization of groundwater in the study area. This analysis defines the main factors whose correlation with the variables allows an explanation of the phenomena involved.

Table 1 shows that electrical conductivity (EC) is significantly correlated with TDS (0.97), with chlorides (0.84), with Sodium (0.62) and Potassium (0.59). TDS is strongly correlated with Chloride (0.83), Sodium (0.59) and Potassium (0.57). Sulfates are strongly correlated to calcium (0.66). Chlorides are strongly correlated to Sodium (0.75) and Potassium (0.82). Bicarbonates are strongly correlated to sulfates (0.53).

Table 2 shows that the analysis of the PCA results also made it possible to calculate the eigenvalues and expressed variances for each factor and their accumulation (Table 1).

Table 1. Correlation matrix of physicochemical parameters of groundwater in the Mellagou Valley

Variables	T°C	pH	e25°C	TDS	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	HCO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻
T°C	1											
pH	0.049	1										
e25°C	0.266	-0.377	1									
TDS	0.258	-0.481	0.972	1								
Ca ²⁺	0.042	-0.149	-0.012	0.032	1							
Mg ²⁺	-0.326	0.089	0.044	0.006	0.179	1						
Na ⁺	0.264	-0.089	0.627	0.592	-0.032	-0.027	1					
K ⁺	0.043	-0.082	0.599	0.568	0.012	0.032	0.824	1				
HCO ₃ ⁻	-0.297	-0.347	0.352	0.476	0.252	0.055	0.408	0.479	1			
Cl ⁻	0.447	-0.270	0.840	0.838	0.100	0.080	0.756	0.778	0.339	1		
SO ₄ ²⁻	0.056	-0.365	0.126	0.251	0.668	0.193	0.007	0.000	0.535	0.185	1	
NO ₃ ⁻	-0.304	-0.079	0.109	0.135	-0.140	0.205	0.065	-0.111	0.017	-0.079	-0.068	1

"Pearson correlation coefficient" (Bold numbers indicate significant correlations)

Table 2. Eigenvalues and percentages expressed by the principal axes

	F1	F2	F3
Eigenvalue	4.560	2.334	1.632
% Variability	38.003	19.453	13.600
cumulative%	38.003	57.456	71.056

The percentage of variances expressed is 38% for factor 1; 19.45% for factor F2. The representation using these two factors gives a satisfactory account of the structure of the scatterplots.

In the space of variables, the F1 axis (57.46%) appears as a mineralization factor on which most of the elements (EC, TDS, Cl^- , Ca^{2+} , Mg^{2+} , SO_4^{2-} , HCO_3^- and NO_3^-) are positively correlated. It explains 38% of the total variance. Note the significant correlation between EC, TDS, Na^+ and Cl^- (Figure 7).

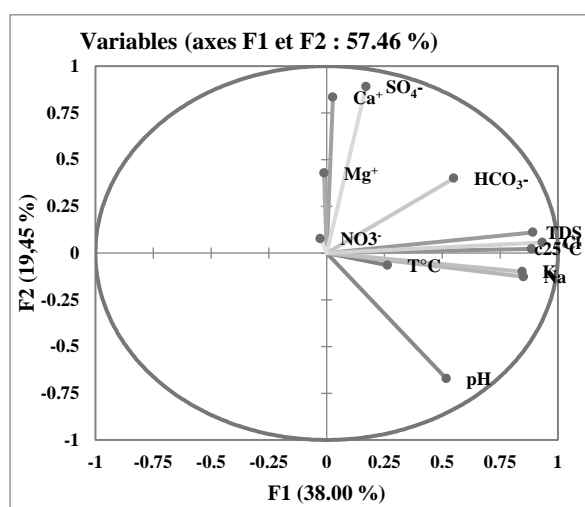


Figure 7. Factorial plan of the Principal Component Analysis (PCA): F1– F2.

The factorial axis F2, expresses 19.45% of the variance, and shows the opposition of two poles, a sodium chloride, sulfate-calcium, and magnesium pole and a bicarbonate-calcium pole. This allows us to distinguish the waters of the superficial Plio-Quaternary aquifer with rapid infiltration, rich in HCO_3^- and Ca^{2+} , and the waters of the deep Cretaceous aquifer rich in Cl^- , Na^+ , SO_4^{2-} and Mg^{2+} , with a higher mineral load, which have remained for a long time in the underground circuit and with slow transit. This is explained by the aridity of the climate, in this semi-arid area, the increase of evapotranspiration can lead to the salinization of groundwater. The presence of nitrates is related to lithological heterogeneity and agricultural inputs. (Drouiche, 2022; Li & Zhang, 2008).

The PCA shows that the main factor influencing the composition of major ions in the watershed is the increase in minerality under the influence of climate, which is usual in North Africa.

The second process is nitrogen contamination, which highlights the impact of agricultural activity on groundwater quality in this region.

4. CONCLUSION

This study contributed to the knowledge of the hydrogeochemical functioning of the Mellagou Aquifer using geochemical tracers and principal component analysis. The waters of the shallow Plio-Quaternary aquifer have calcic bicarbonate facies. This water becomes concentrated with gypsum salt elements along its pathway and transforms into sodium chloride to calcium sulfate facies.

Several processes are expected to control the geochemistry of this aquifer. These include dissolution, precipitation of mineral salts, cation exchange, and the influence of climate, which increases the minerality of water, but also agricultural pollution. The saturation indices allowed us to know that all the evaporite elements are undersaturated.

According to the calcite SI and dolomite SI values, the majority of the sampled bridges are supersaturated, therefore precipitate. Water infiltrated at the Cretaceous limestone level of the deep aquifer is enriched in calcium and bicarbonates, along its underground path and in the presence of dolomite, and gypsum, halite, it results in water enriched mainly in sulfates, sodium and chloride. This clearly indicates a dominant geological factor in the enrichment of these elements.

The PCA confirmed the hypothesis of the origin of the mineralization and the acquisition of the saline load. These hydrochemical evolution mechanisms are largely similar to those delineated for similar regional aquifer systems in Algeria and around the world. These results will be a basis for monitoring and protecting the water resources of this watershed.

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