

Geochemical and multivariate statistical study to assess the salinisation origin of the Remila plain groundwater, Khenchela Algeria

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Abstract: The aquifer system of the Remila plain (Khenchela, Algeria), covering 250 km², is one of the semiarid regions where groundwater is heavily exploited for urban supply and irrigation. An integrated hydrochemical and statistical analysis was performed on 70 water samples to identify the main processes and the origin of salinisation of our waters. Chemical analyses indicate salinity values (*TDS*) ranging from 568 to 1586 mg·dm⁻³ with an average of 869 mg·dm⁻³, with sulphate being the dominant ions, especially in the north and northeastern parts of the region. The identified chemical facies are SO₄-Cl-Ca in the northeastern part, SO₄-Cl-Ca-Mg present in most waters, and HCO₃-Ca-Mg in the southeastern part.

We applied the statistical approach to group the waters into three categories using Principal Component Analysis (PCA) and Hierarchical Clustering Analysis (HCA); 1) saline waters (23%) (*TDS* > 1000 mg·dm⁻³ and SO₄²⁻ dominance), 2) moderately saline waters (51%) with HCO₃⁻ dominance, 3) moderately saline waters (26%) with a mixed facies. The binary ion diagrams used suggest that the main hydrochemical processes are: evaporites dissolution and/or precipitation, accompanied by an exchange and/or reverse exchange of ions. Additionally, another process was detected in the northeastern part of the area; the saline intrusion of Sabkha waters, favoured by intensive aquifer exploitation.

Keywords: groundwater, hydrogeochemistry, salinity, statistics, Remila

INTRODUCTION

Freshwater resources account for no more than 3% of the planet's water and are mostly in frozen form, where groundwater accounts for only 30% of the world's freshwater [BENCHOKROUN 2008; CASSARDO, JONES 2011]. Groundwater has become an important source of water supply for the domestic, industrial, and agricultural sectors in many countries, especially in arid and semi-arid regions [AOUIDANE 2017; BOUZOURRA *et al.* 2015; PEJMAN *et al.* 2009]. In Algeria, 67% of irrigated lands are irrigated by groundwater [ZEKTSER, EVERETT (eds.) 2004]. However, water quality plays an important role in plant growth, human health,

and the environment. Natural processes can cause water quality to deteriorate and some of these processes are exacerbated by human activities, due to the over-exploitation of groundwater resources [MASOUD *et al.* 2018; RINA *et al.* 2013]. The identification of geochemical processes in groundwater is, therefore, crucial for the sustainable management of these resources.

Salinisation is one of the most common contaminants in water resources, occurring particularly in groundwater in arid and semi-arid regions [PISINARAS *et al.* 2010]. Over the last few decades, salinisation of groundwater has been the subject of extensive research [AOUIDANE, BELHAMRA 2017; GIBBS 1970; PAZAND *et al.* 2012]. This problem can be attributed to natural and/or

anthropogenic factors, such as the interaction of water with rocks [FRAPE *et al.* 1984; GHESQUIÈRE *et al.* 2015], the intrusion of coastal saltwater or Sabkha water generated by excessive groundwater pumping [CAPACCIONIA *et al.* 2005], fossil seawater, which is a primary source of Cretaceous seawater [FARID *et al.* 2013; TJJANI 2004]. Finally, the anthropogenic intervention compounded local natural processes by saline water intrusion, due to over-exploitation of groundwater, inadequate sewage disposal, intensive agricultural and industrial activities [ZEWDU *et al.* 2017]. In general, many approaches can be used to identify groundwater salinisation processes. These include geochemical analysis, using binary ratios of major and minor tracers [GIL-MÁRQUEZA *et al.* 2017; SANCHEZ-MARTOS *et al.* 2002], using isotopic tracers [GHABAYEN *et al.* 2006; HASSEN *et al.* 2016], and performing statistical analysis [KHARROUBI *et al.* 2012; MASOUD *et al.* 2018; SUDHEER *et al.* 2017; VISWANATH *et al.* 2015].

Like many arid and semi-arid zones, the Remila endorheic basin presents this problem, which is the subject of this study. It is situated in northeastern Algeria and is limited to the north by the depression of Sabkha Garaat Etarf. The area is known for its predominantly agricultural character with a high population, and the water supply comes mainly from groundwater. Therefore, it is important to understand the main processes that control the salinisation of these waters, within the limits of improving water resource quality management in this region.

The main aim of this study is to identify factors that control groundwater salinity. An integrated approach involving geological, hydrogeological, hydrochemical, and statistical methods was used to (1) assess groundwater chemistry to highlight dominant groundwater mineralisation processes, (2) use statistical analyses to identify distinct water groups and assess the correlation between different elements and factors of water hydrochemistry, (3) and identify recharge areas and major sources of contamination. A better understanding of the sources of salinisation and contamination of the Remila aquifer system (Khenchela) will then serve as an important scientific basis for the water resource planning and management in the study area.

MATERIALS AND METHODS

STUDY AREA

The study area is part of the endorheic basin of Garaat Et Tarf. It is located in northeastern Algeria and lies between the parallels 35°25' and 35°40' north latitude, and 06°30' and 07°05' east longitude, on an area of 250 km² with flat topography and an altitude of 800 to 1000 m. The study area is surrounded to the south by the northern slope of the Aurès Mountains, which is the recharge area; to the north by the Djbel Fdjoudj mountain range, and to the northeast by the depression Sabkha of Garaat Et Tarf (Fig. 1) [CPH 1977]. The semi-arid climate of the region is characterised by annual precipitation of less than 400 mm with an average annual temperature of 17°C. The minimum and maximum temperatures were noted in January (6°C) and in August (30°C), respectively. The potential annual evapotranspiration is 1050 mm. Surface water in the area is scarce, and groundwater is the only source of water for urban and agricultural use.

Geological setting

From the geological point of view, the Remila region is located in a depression zone which is 50 to 60 km long and c.a. 20 km wide. It represents a floodplain of Mio-Pliocene with quaternary deposits transported by streams and then deposited in a sequence of floodplains due to stream flow regimes [CGG 1969; LAFFITTE 1939]. The Cretaceous series surrounding the study area is composed of Middle Cretaceous (Aptian) or Upper Cretaceous (Cenomanian, Turonian, and Senonian), which are the most extensive on the southern and northern side. While in the northwest part, the Miocene is represented by a limestone-sandstone and limestone deposit (Burdigalian) arranged discordantly on the Cretaceous and the red Pontian marl. An endorheic saline depression (Sabkha Great Etarf) of 200 km² occupies the eastern part of the plain [VILA 1977]. The study area's lithostratigraphy (Remila plain) is characterised by the development of recent Quaternary soils (Fig. 1).

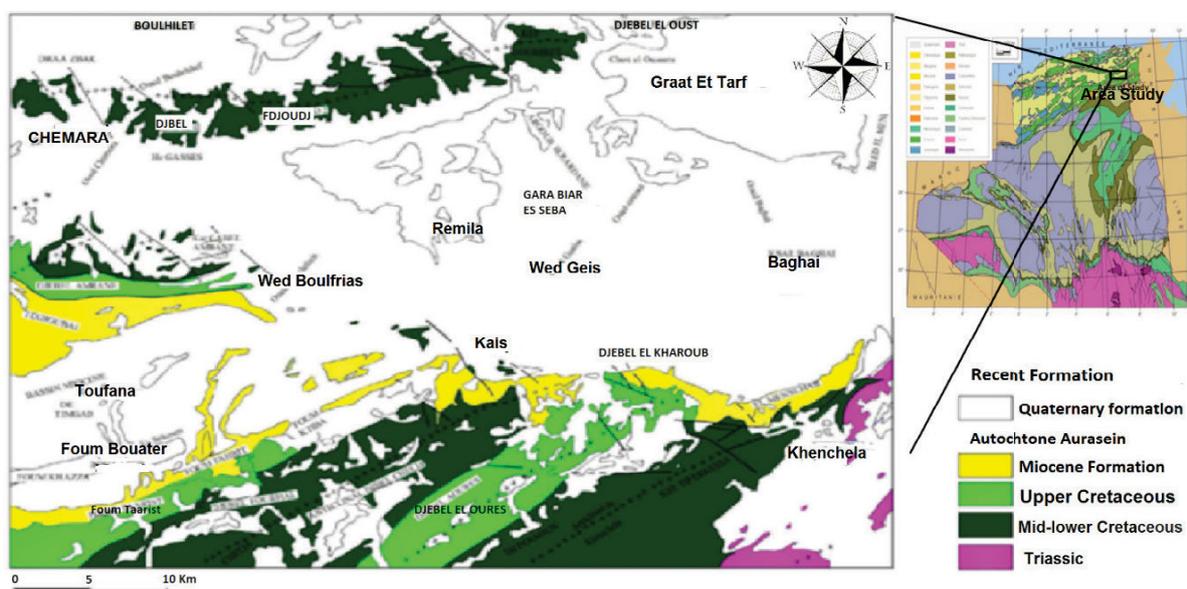


Fig. 1. Map showing location and geological formations of the study area; source: VILA [1977]

Hydrogeological setting

The distribution of the hydrographic system in the study area is related to the evolution of the structural phenomena. The hydrographic network is characterised by a high density with a temporary flow during the wet seasons. The Boulefreis Oued drains the entire area from its source in the southwest to discharge to the Sabkha in the northeast. Also, there are other less important streams; the Baghai Oued, draining the eastern part, while the centre of the area is drained by Maarouf Oueds and Gueiss Oued. The flow of these streams is temporary and pours into a closed depression (Sabkha of Garaat Etarf) [DHWK 1984].

The Remila aquifer lies in a sandy-marly limestone substratum of the Moi-Plio-Quaternary. The geochemical properties and groundwater recharge mode will generally depend on the aquifer's depth and lithostratigraphy. Aquifer recharge is maintained through the drainage of runoff water from the northern slope of Aures. The MPQ aquifer is represented by two water tables, located in the alluvial filling of the plain [CGG 1969].

SAMPLE COLLECTION

Groundwater samples were collected during two campaigns, from 35 boreholes, and were used to identify the dominant hydro-geochemical processes in the Remila aquifer system. The first campaign was carried out in the dry period in December–October 2013 and the second was carried out in the wet period of May–June 2014. We used the methods recommended by RODIER *et al.* [2016] in the sampling and analysis to minimise handling errors. The borehole water was pumped out two to three times before sampling. Regular monitoring of the electrical conductivity was performed at the same time until the values were stable.

The samples taken from the water were untreated and raw. For each sample, water was taken from a 1 dm³ pre-rinsed polyethylene bottle and filtered through a 0.45 µm membrane filter. The bottles were filled to minimise exposure to air, and transported in coolers at 4°C. The sampling point coordinates were determined using a global geographic positioning system (GPS GARMIN Olathe KS, US) (Fig. 2).

SAMPLE ANALYSIS

The physicochemical parameters of the water, such as electrical conductivity (*EC*), total dissolved solids (*TDS*), pH, and temperature were measured in situ using a Consort C535 multiparameter with specific probes, which had been pre-calibrated in the laboratory. Also, the measurement of the other parameters was conducted at Constantine University, Algeria, in the hydrochemical laboratory (Tab. 1).

Table 1. Methods used for major ion and isotope analysis in the waters of Remila

Element	Method of analysis
Ca ²⁺ and Na ⁺	industrial flame photometers PFP7
Mg ²⁺ , K ⁺ , Sr ²⁺ , Al ³⁺ and Li ⁺	Perkin Elmer atomic absorption AA 200 (FXAA)
SO ₄ ²⁻ and NO ₃ ⁻	colorimetric method
Cl ⁻ and HCO ₃ ⁻	volumetric method

Source: own elaboration

Multivariate statistical analysis of the data was one of the methods used to analyse geochemical data, allowing the large dataset of variables to be reduced to a few factors, so as to elucidate correlations between chemical parameters. Our statistical analysis of the data was carried out using STATISTICA 7.1 (StatSoft1) software. Principal Component Analysis (PCA), Correlation Table, and Hierarchical Clustering Analysis (HCA) were applied in our case.

RESULTS AND DISCUSSION

GENERAL HYDROCHEMICAL FEATURES

Spatial distribution of groundwater salinity

The results of the groundwater physicochemical analysis are presented in Table 2. Salinity is one of the most important water-classification parameters. Water salinity values (*TDS*) ranging

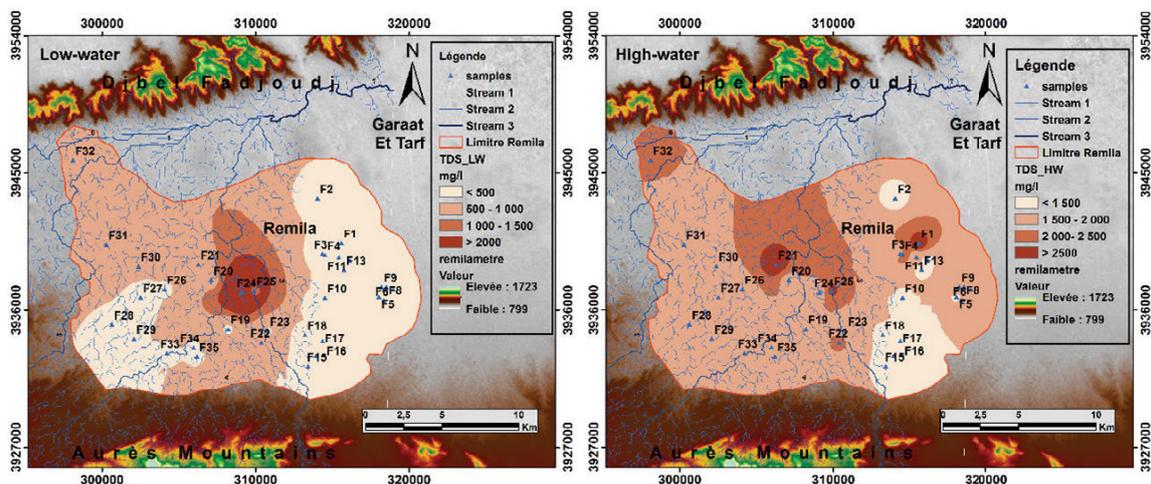


Fig. 2. Spatial salinity distribution map (*TDS*) of groundwater Remila (Khenchela); source: own study

from 568 to 1586 mg·dm⁻³, with an average of 906.3 mg·dm⁻³, indicate low to medium mineralisation of water during the dry season. While the TDS values in the wet season ranged from 1250 to 3270 mg·dm⁻³, with an average of 1822.16 mg·dm⁻³, the mineralisation of our waters increased from medium to high. After the wet season we notice a significant increase in the salt content. It suggests that the rainwater that feeds the groundwater influences the mineralisation of our waters. While the pH values of most of the points are close to neutrality where they are between 6.5 and 7, with an average of 6.9 during the wet period, and values oscillating between 6.5 and 7.21, with an average of 6.91 during the dry period, seasonal variation is slight.

The results obtained for the 35 points were used to generate a spatial distribution map of salinity (TDS) (Fig. 2). We noted that the piezometric map of groundwater presents a rather close resemblance, reflecting changes in the mineralisation of the water, upstream to downstream (south to north), although some specific trends are also apparent. This spatial variation is classifiable into two water types: the waters of the southern and western parts of the plain are low in salt, while the waters of the northern part near the Sabkha are moderately salty. This is possibly due to the strong influence of Sabkha's salt waters.

Hydrochemical facies

Identifying the hydrochemical type of water is a useful tool for assessing water chemistry and processes such as mixing, cation exchange, and dissolution that affect the composition of groundwater. The Piper diagram [PIPER 1944] is considered to be

a popular method of performing multiple analyses on the same graph. It is useful for grouping samples and defining the various groundwater origins [WALTON 1965]. The main ionic species obtained from the geochemical analysis of 70 samples are projected on the Piper diagram using DIAGRAMS 5.1 software (Fig. 3). Based on these results, we deduced the dominance of three chemical facies for the two campaigns (dry period and wet period) in our waters: SO₄-Cl-Ca saltwater represented by certain points in the north-eastern part of the area near Sabkha, SO₄-Cl-Ca-Mg moderately saline waters represented by the majority of points, and the good quality type HCO₃-Ca-Mg represented by some points in the southeastern part of the area.

Most of the water points show a tendency for SO₄²⁻ dominance in the anionic triangle, which applies especially to samples collected after the wet season, while in the cationic triangle there is a tendency towards Ca²⁺ domination. This suggests that the aquifer is affected by different hydrochemical processes: ionic exchange with the substratum, leaching and dissolution of sediments in the southern part (recharge areas after a wet season), and saltwater intrusion of the Sabkha waters in the northeastern regions.

STATISTICAL ANALYSIS

A descriptive statistical evaluation of data is a critical step in any statistical analysis (Tab. 2) [HOSSEINI *et al.* 2014; YANG *et al.* 2011]. The measured parameter values are characterised by very high variance. The most important factor that can describe the

Table 2. Descriptive statistics of Remila water parameters (mg·dm⁻³)

Parameter	Campaign	Mean	Median	Minimum	Maximum	SD	CV	Skewness	Kurtosis
pH	dry period	6.87	6.92	6.52	7.02	0.13	1.90	-1.51	1.33
	wet period	6.91	6.92	6.50	7.21	0.11	1.55	-1.27	7.09
TDS	dry period	896.29	824.50	568.00	1586.00	254.52	28.40	1.29	1.13
	wet period	1797.14	1710.00	1250.00	3270.00	485.57	27.02	1.67	2.87
Ca ²⁺	dry period	134.74	132.26	40.08	252.50	39.36	29.21	0.49	2.41
	wet period	132.14	128.26	44.09	248.50	46.42	35.13	0.76	1.18
Mg ²⁺	dry period	50.83	35.88	6.96	239.64	53.54	105.34	2.63	6.50
	wet period	56.83	52.68	28.80	129.48	22.48	39.55	2.04	4.59
Na ⁺	dry period	55.56	41.75	11.96	145.59	34.78	62.61	1.45	1.49
	wet period	69.46	62.11	29.67	162.15	33.45	48.16	0.93	0.41
K ⁺	dry period	1.09	0.17	0.02	9.98	2.09	191.45	2.94	9.57
	wet period	1.16	1.00	0.09	3.95	0.80	69.13	1.87	4.43
HCO ₃ ⁻	dry period	174.75	164.70	54.90	280.60	53.64	30.70	0.31	-0.30
	wet period	253.20	256.20	183.00	402.60	48.86	19.30	0.96	1.39
Cl ⁻	dry period	122.80	118.93	35.50	312.40	58.36	47.53	1.16	2.07
	wet period	125.79	117.15	35.50	333.70	83.41	66.31	1.22	0.90
SO ₄ ²⁻	dry period	344.24	320.00	160.00	704.00	129.92	37.74	1.64	2.84
	wet period	293.86	270.00	140.00	1250.00	188.44	64.13	4.05	20.20
NO ₃ ⁻	dry period	-	-	-	-	-	-	-	-
	wet period	8.19	3.60	0.20	39.00	11.09	135.45	1.96	2.42

Explanations: SD = standard deviation; CV = coefficient of variation.

Source: own study.

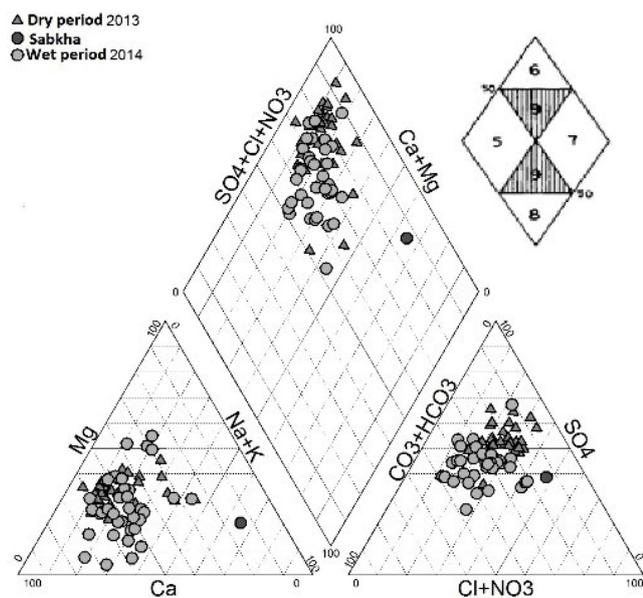


Fig. 3. Piper diagram of Remila groundwater (dry period and wet period); source: own study

variability of the water parameter values is the coefficient variance (CV). A low coefficient variance (<10%) for pH was obtained suggesting a low spatial variability in the study area. While for the TDS, HCO₃⁻, Ca²⁺, and Fe²⁺, a moderate coefficient variance (20–50%) was observed, it indicates moderate spatial variability for these parameters and is mainly influenced by natural factors. Nonetheless, for Mg²⁺, Na⁺, K⁺, SO₄²⁻, Cl⁻ and NO₃⁻, very high CV values exceeding 50% were observed, suggesting a very high spatial variability. These parameters are affected by natural and anthropogenic factors.

Skewness reflects the symmetric distribution when these values are lower than 0 and the totality of the data is below the average. When data is normally distributed, the skewness value equals zero [BROVELLI *et al.* 2011; GALVAO *et al.* 2013]. If the

calculated skewness values of Ca²⁺, Na⁺, and HCO₃⁻ are close to zero, it indicates that the data follow a normal distribution. However, when the other parameters have values >1, this does not require a data transformation. OLIVER *et al.* [2002], KERRY and OLIVIER [2007], and OLIVER [2010] have shown that values of skewness outside the standard limit (±1), do not necessarily imply the need to transform data into statistics for further study, especially when the data values were high.

Pearson’s correlation matrix

Many significant correlations between the different ions were established using the correlation matrix of the various parameters (Tab. 3). A strong correlation ($r > 0.7$) was found between the chemical tracers SO₄²⁻, Cl⁻, Mg²⁺, Na⁺, Ca²⁺, Sr²⁺ and total dissolved solids (TDS), indicating an evaporite origin (from NaCl, CaSO₄, CaSO₄ 2H₂O, MgSO₄ or SrSO₄) of these elements in the water. However, this is not the case for HCO₃⁻ which has a low correlation with the salinity indicator TDS. This is explained by its low concentration in the water solution that ranges between 54.9 and 280.6 mg·dm⁻³. Moreover, the correlation coefficients between chloride ($r = 0.81$) and sodium ($r = 0.81$), with the TDS, suggests the same origin for these two elements. On the other hand, there is a slight difference between sulphate ($r = 0.95$), magnesium ($r = 0.87$), and calcium ($r = 0.66$) correlation coefficients with TDS, which can be explained by the unconservative transport of calcium and magnesium ions (cation exchange with clay minerals which are most abundant in the area and/or participate in precipitation/dissolution reactions) [VAN BREUKELLEN *et al.* 1998].

As far as the relationship between anions and cations is concerned, significant correlations were found between Cl⁻ and Na⁺ ($r = 0.55$), Cl⁻ and Ca²⁺ ($r = 0.74$), Cl⁻ and Mg²⁺ ($r = 0.68$), Cl⁻ and K⁺ ($r = 0.65$), SO₄²⁻ and Mg²⁺ ($r = 0.93$), SO₄²⁻ and Ca²⁺ ($r = 0.61$). This tends to prove that most chloride comes from the dissolution of KCl and NaCl but a very small proportion of this anion could come from the dissolution of other minerals. The low correlation between chloride and sodium can be explained by the

Table 3. Correlation matrix of different groundwater parameters (mg·dm⁻³)

Variable	pH	TDS	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	HCO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	Fe ²⁺	Mn ²⁺	Sr ²⁺
pH	1.00											
TDS	0.16	1.00										
Ca ²⁺	0.22	0.66	1.00									
Mg ²⁺	0.26	0.87	0.43	1.00								
Na ⁺	0.00	0.81	0.17	0.69	1.00							
K ⁺	0.16	0.82	0.38	0.79	0.75	1.00						
HCO ₃ ⁻	-0.33	0.20	-0.14	-0.02	0.44	0.14	1.00					
Cl ⁻	0.31	0.81	0.74	0.68	0.55	0.65	-0.18	1.00				
SO ₄ ²⁻	0.19	0.96	0.61	0.93	0.74	0.80	0.02	0.73	1.00			
Fe ²⁺	-0.08	0.21	0.28	0.15	0.00	0.20	0.08	0.16	0.20	1.00		
Mn ²⁺	0.01	-0.24	-0.36	-0.21	-0.05	-0.12	0.24	-0.44	-0.22	-0.07	1.00	
Sr ²⁺	-0.01	0.71	0.58	0.57	0.45	0.62	0.34	0.46	0.66	0.36	-0.04	1.00

Source: own study.

involvement of sodium ions in cation exchange with the clay substratum of the aquifer. Additionally, there was a relatively significant correlation between Cl^- with Ca^{2+} (0.74) and Cl^- with Mg^{2+} (0.68). This is due to the water salinity that is caused by several processes that characterise the highly mineralised waters, including an exchange of cations and dissolution of gypsum and halite that can increase the concentrations of calcium, magnesium, and chloride, respectively.

We carried out the principal component analysis (PCA) and hierarchical clustering analysis (HCA) using the software STATISTICA 8 in an attempt to clarify the relationship between the chemical elements (variables) and grouping individual water points with the same chemistry.

Principal component analysis (PCA)

More and more studies are based on statistical analysis to define water types; one of the most recommended approaches is the principal component analysis [AYADI *et al.* 2018; FARNHAM *et al.* 2003 in ATTEIA 2015]. In the Remila aquifer samples, the variable factors F1–F2 (Fig. 4a, Tab. 4) show that this plan accounts for 53.95% of the expressed variance. The factor F1 (36.74%) is negatively determined by most of the elements: TDS , SO_4^{2-} , Cl^- , Mg^{2+} , K^+ , Na^+ , and Ca^{2+} and therefore has a mineralisation axis of evaporites and salinity. A very good correlation exists between TDS , SO_4^{2-} and Mg^{2+} . The F2 explains 17.21% of the total variance, which is positively associated with pH and HCO_3^- ,

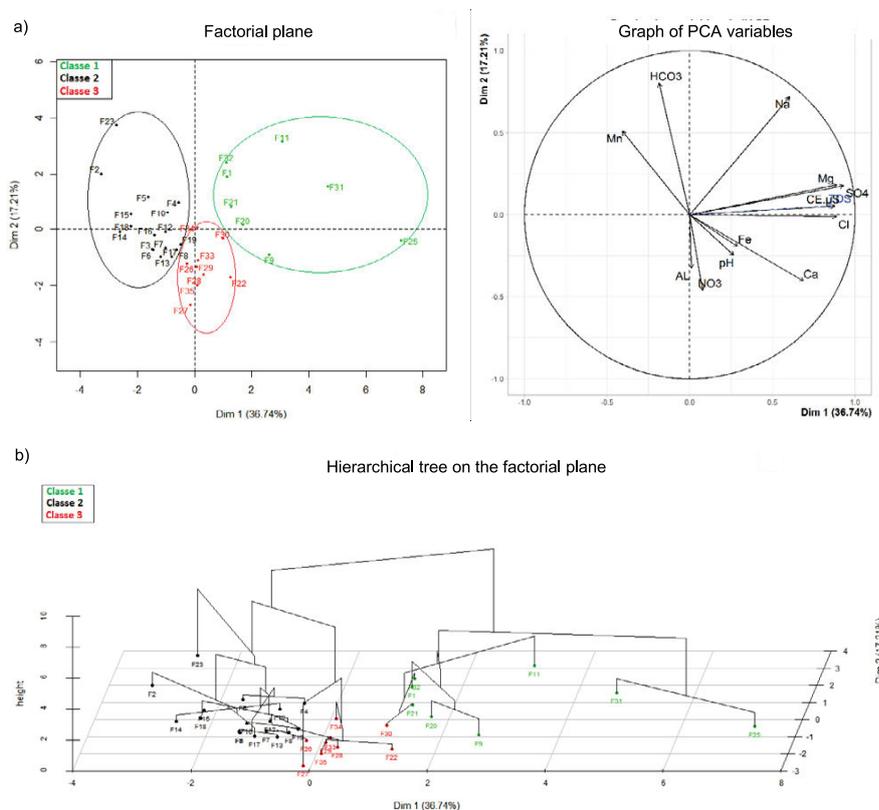


Fig. 4. Statistical analysis: a) Principal Component Analysis, b) Ascending Hierarchical Clustering of Remila waters (Khenchela); source: own study

Table 4. Varimax factor loading matrix, communalities for each variable analysed

Parameters	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5
pH	-0.2245	-0.5564	0.4408	0.1886	-0.2360
TDS	-0.9795	0.0831	0.0054	-0.0857	0.0711
Ca^{2+}	-0.6711	-0.3059	-0.4608	0.0097	-0.2072
Mg^{2+}	-0.8954	-0.0599	0.2170	0.0042	0.0861
Na^+	-0.7563	0.3892	0.3325	-0.1522	0.2471
K^+	-0.8628	0.1267	0.1683	-0.0418	0.0688
HCO_3^{2-}	-0.0889	0.8501	-0.0524	-0.0639	0.2252
Cl^-	-0.8338	-0.3110	-0.0842	-0.0693	-0.0157
SO_4^{2-}	-0.9552	-0.0205	0.0742	-0.0525	0.0339
Fe^{2+}	-0.2704	0.0499	-0.5723	0.5659	0.1012
Mn^{2+}	0.2749	0.4881	0.4029	0.2935	-0.3649
Sr^{2+}	-0.7282	0.3298	-0.3126	0.1030	-0.1388

Source: own study.

suggesting that the dissolution of carbonates does not contribute to the mineralisation of water.

The projection results variables and individuals in the factorial plan (F1, F2) shown in Fig. 4a, identified three different water groups. The first type (I) are saline waters representing 23% of the total, with *TDS* exceeding $1000 \text{ mg}\cdot\text{dm}^{-3}$ and characterised by a high concentration of SO_4^{2-} . Type (II) are non-saline water representing 51% of the total concentrations of HCO_3^- and type (III) is moderately saline waters with mixed facies present in the eastern part of the region and making up 26% of the total.

Hierarchical cluster analysis (HCA)

The Ascending Hierarchical Clustering (AHC) is a powerful multivariate statistical technique to analyse water chemistry data for geochemical model formulation [AHOUSI *et al.* 2010; YIDANA 2008].

The results from the analysis of the hierarchical ascending data classification can be observed in the dendrogram using the Euclidean distance for similarity measures (Fig. 4b), which highlights three principal groupings of the variables. The first group includes individual water points located in the north-western part of the study area, it contains salty waters characterised by a dominance of SO_4^{2+} . The second group represents most waters in the eastern and south-eastern parts of the region, characterised by low salinity with a mixed face. The last group includes the waters of the south-western part that are moderately saline with a dominance of HCO_3^- .

The results provided by the PCA of the individual samples and ascending hierarchical clustering are in perfect agreement since they demonstrated that water mineralisation is dominated by sulphate. Besides, the typology of the water samples can be represented by three facies.

GROUNDWATER MINERALISATION PROCESSES

Correlations show a statistical relationship between two variables or more. Our results confirm a significant correlation between SO_4^{2-} , Mg^{2+} , Cl^- , Ca^{2+} , Na^+ , HCO_3^- , Sr^{2+} , K^+ , and *TDS*, indicate the contribution of these elements to the salinity of the groundwater, and suggest that the dissolution of evaporitic rocks may be the common source of mineralisation. Such results require geochemical calculations of speciation-solubility in order to identify the dominant processes, which influence the hydro-geochemistry of the Remila plain groundwater.

Water-rock interaction

To assess the water-rock interaction processes, diagrams of ion ratio of $\text{Ca}^{2+}/\text{Na}^+$ versus $\text{HCO}_3^-/\text{Na}^+$ and $\text{Ca}^{2+}/\text{Na}^+$ versus $\text{Mg}^{2+}/\text{Na}^+$ were used to identify the influence of silicate weathering, dissolution evaporites, and carbonate alteration on mineralisation of groundwater in the study area. The projection results of the water points (Fig. 5a) imply that the bulk of the waters is affected by silicate weathering with a bent towards carbonate dissolution, especially after the wet period. The spatial distribution suggests that the waters near the limestone formations of the eastern part are influenced by carbonate dissolution, whereas the waters of the northwestern part are affected by silicate alteration. The normalisation of Na by intake Ca versus Mg (Fig. 5b), indicates that the bulk of Mg within the waters of the eastern part is released by carbonate dissolution, in contrast, the Mg within the waters of the western part are influenced by silicate weathering. The $\text{Ca}^{2+}/\text{Mg}^{2+}$ ionic ratio is widely used to identify the dissolution of calcite and dolomite. When the ratio values are ≤ 1 , it indicates the dominance of the dissolution of dolomite; if the values are between 1 and 2 it indicates the dominance of the dissolution of calcite; and if the values are ≥ 2 , waters are affected by silicate minerals [SINGH *et al.* 2017; SRINIVASAMOORTHY *et al.* 2014]. The ratio calculation outcomes suggest that the waters in the extreme south are dominated by dolomite dissolution, the majority of the waters in the primary central part of the area are dominated by calcite dissolution, whilst the northern part is affected by silicate weathering.

The Gibbs diagrams [GIBBS 1970] are widely used to assess the functional source of ions dissolved in water such as the dominance of precipitation, weathering of rocks, and dominance of evaporation which controls the water chemistry [MASOUD *et al.* 2018; SINGH *et al.* 2011; VAROL, DAVRAZ 2014]. The chemical data are plotted in a semi-logarithmic dispersion of *TDS* values versus anion ($\text{Cl}^-/(\text{Cl}^-+\text{HCO}_3^-)$) and cations ($\text{Na}^+/(Ca^{2+}+Na^+)$) ratios, where all concentrations of ionic values are expressed in $\text{meq}\cdot\text{dm}^{-3}$ (Fig. 6a, b).

As shown by the data projected in the Gibbs diagrams, most of the groundwater samples of the two seasons are located in the predominantly rocky zone, but also have a tendency towards evaporation and precipitation. The mineralisation of our waters is therefore controlled by the dissolution of evaporitic rocks and is also affected by the waters of Sabkha, which are exposed to intense evaporation with chemical precipitation of minerals, which increases the salinisation of our waters, particularly in the northeastern part of the area.

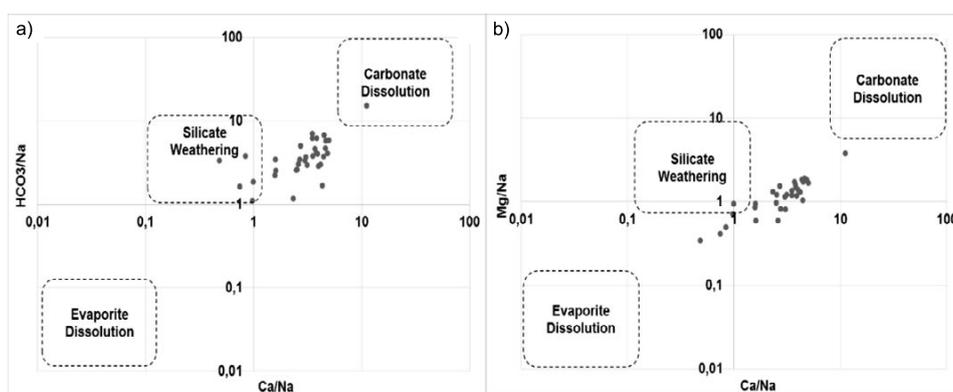


Fig. 5. Scatter plots of: a) $\text{Ca}^{2+}/\text{Na}^+$ versus $\text{HCO}_3^-/\text{Na}^+$, b) $\text{Ca}^{2+}/\text{Na}^+$ versus $\text{Mg}^{2+}/\text{Na}^+$ ratio of the Remila groundwater samples; source: own study

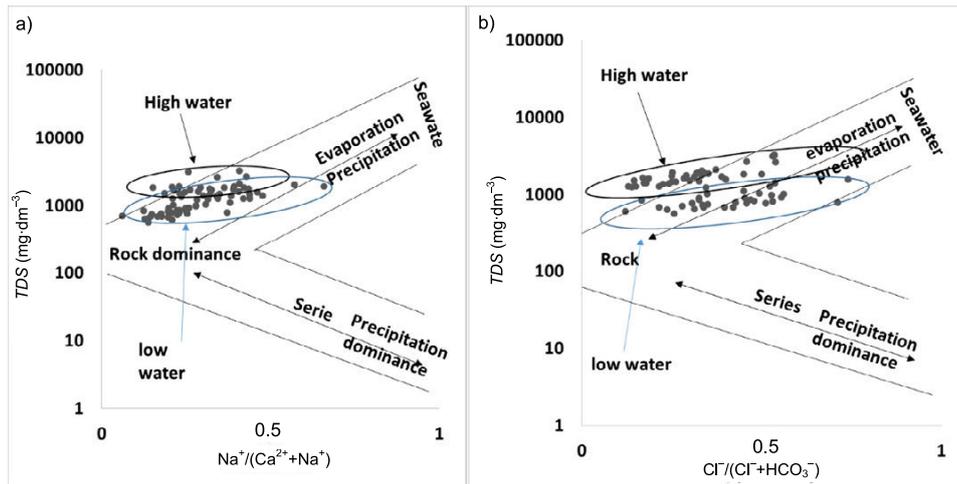


Fig. 6. Gibbs diagrams of Remila groundwater; source: own study

Cationic exchange

Most of the results show an excess concentration of cations by anion supply, which indicates that another source of these cations, such as exchange, should be possible. Nevertheless, SCHOELLER’s [1965] proposed chloroalkaline index (CAI) can be used to determine the degree of ion exchange reactions between the aquifer substratum and groundwater. It is a commonly used tool for recognition of dominant processes of ion exchange in groundwater [AYADI *et al.* 2018]. The CAI is calculated using the equation $(Cl^- - (Na^+ + K^+)) / Cl^-$ where the values are expressed in $meq \cdot dm^{-3}$. If the CAI values tend to decrease (negative), it indicates a dominance of the basic processes of ion exchange, where Ca^{2+} and Mg^{2+} are adsorbed on the substratum and K^+ and Na^+ are released in water. Thus, if the CAI index values tend to increase (positive), this implies a dominance of the processes of reverse ion exchange, where K^+ and Na^+ are adsorbed on the substratum and Ca^{2+} and Mg^{2+} are released in water. Meanwhile, if the CAI values are close to zero/are in balance, this implies the absence of an exchange process.

The calculated chloroalkaline index in the study area shows positive values for most groundwater samples (68%), which indicates the dominance of the reverse exchange process in which Ca^{2+} , Mg^{2+} of the aquifer substratum are released, and Na^+ , K^+ are adsorbed. Although 26% of the samples have values close to zero, which implies equilibrium and indicates an absence of ion exchange, they are found in the southeastern part of the region. Meanwhile, only 2% of the samples show negative values,

indicating the dominance of direct ion exchange processes, releasing Na^+ , K^+ from the aquifer substratum and fixing Ca^{2+} , Mg^{2+} from the water, especially in the northeastern part of the area.

Significant fluctuations in the concentration of major ions occur by various processes, while ion exchange often causes changes or reversals of groundwater cationic concentrations. Studying the relationship between $(Ca^{2+} + Mg^{2+})$ and $(HCO_3^- + SO_4^{2-})$ will allow us to identify the processes that influence groundwater mineralisation. The binary diagram of $(Ca^{2+} + Mg^{2+})$ versus $(HCO_3^- + SO_4^{2-})$ (Fig. 7a) shows a projection of the water points around the line (1:1). Generally, the water points, which are close or on the line (1:1), are under the influence of the dissolution of calcite, dolomite, anhydrite, and gypsum [HAMZAOUI-AZAZA *et al.* 2012; HASSEN *et al.* 2016]. While the water points that exist above the line (1:1), present an excess of $(Ca^{2+} + Mg^{2+})$ that are accompanied by ion exchange [TLILI-ZRELLI *et al.* 2013], the water points located below the line (1:1), have a deficit in $(Ca^{2+} + Mg^{2+})$; this decrease in concentration is due to reverse ion exchange [RINA *et al.* 2013]. The result of the water points projection in the diagram (Fig. 7a) shows that most waters are above line (1:1) due to excess $(Ca^{2+} + Mg^{2+})$ and this indicates that reverse ion exchange is a very abundant geochemical process in the region. The water points located in the northeastern part near Sabkha (saltwater) are projected below the line (1:1), suggesting a deficit in $(Ca^{2+} + Mg^{2+})$, which indicates that the waters in this part of the study area are under the influence of ion exchange and saline intrusion.

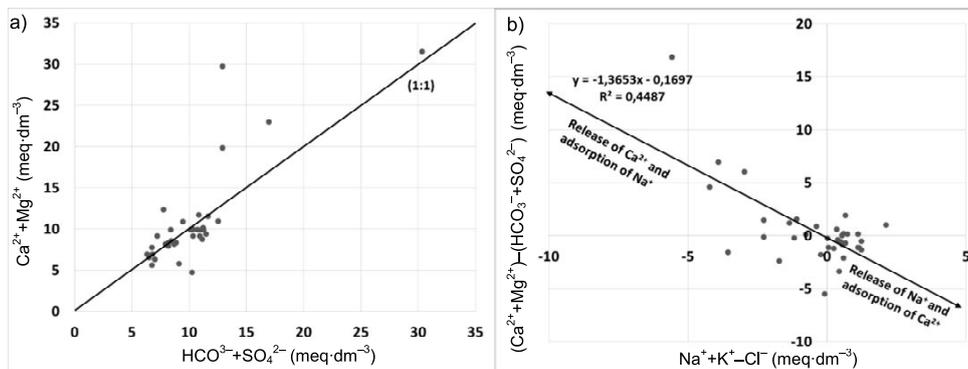


Fig. 7. Scatter plots of: a) $(Ca^{2+} + Mg^{2+})$ versus $(HCO_3^- + SO_4^{2-})$, b) $(Ca^{2+} + Mg^{2+}) - (HCO_3^- + SO_4^{2-})$ versus $(Na^+ + K^+) - Cl^-$ in $(meq \cdot dm^{-3})$ of the Remila groundwater samples; source: own study

For further confirmation that the ion exchange process affects the hydrochemistry of Remila groundwater, another diagram was generated. This diagram has been widely used in various studies [KRAIEM 2015]. The diagram $(Ca^{2+}+Mg^{2+})-(HCO_3^-+SO_4^{2-})$ versus $(Na^++K^+)-Cl^-$ in $(meq\cdot dm^{-3})$ (Fig. 7b) shows that the ion exchange process may be a feature of the study area, since most water points follow a straight line ($R^2 = 0.72$) with a slope of 1.08, suggesting that the cations Ca^{2+} , Mg^{2+} , Na^+ , and K^+ are integrated into the cation exchange reactions. Sample distribution indicates that the most saline groundwater in the western part of the area has migrated in a direction that is characterised by a rise in Ca^{2+} and Mg^{2+} and a decrease in Na^+ and K^+ concentrations, which suggests that the water mineralisation is affected by rock dissolution accompanied by ion exchange. Although water points located in the northeast of the area are characterised by a high concentration of sulphate, they show a slight decrease in Ca^{2+} and Mg^{2+} , whereas significant increases in Na^+ and K^+ are observed. Such findings can be explained by the effect of reverse ion exchange, in which Na^+ is released from the substratum and the Ca^{2+} is adsorbed, and this is favoured by the saline intrusion of Sabkha waters [APPELO 1994]. These results confirm that two main processes control our water hydrochemistry: the dissolution of evaporitic rocks, and ion exchange.

CONCLUSIONS

Salinisation of water is considered a major problem related to land degradation in arid and semi-arid regions. Our research focuses on the endorheic basin of Remila (Khenchela), which is characterised by a semi-arid climate with low and sporadic rainfall. In the study area, the principal source of drinking and irrigation water is groundwater (Plio-quaternary aquifer). A hydrogeochemical and statistical approach was applied in the context of identifying the various salinisation processes and the origin of degradation of the aquifer system in the study area.

The results indicate the predominance of three groundwater facies; SO_4 -Cl-Ca-Ca salt water located to the northeast of the area, SO_4 -Cl-Ca-Mg moderately saline water which is present in most water points, and the type of good quality HCO_3 -Ca-Mg found in the south-eastern part of the area. The statistical analysis of the measured parameters reveals a normal distribution of the data, and the CV indicates a moderate to high spatial variability, which suggests that the parameters measured are mainly affected by environmental factors and by anthropogenic activities. Significant correlations ($r > 0.7$) between the elements SO_4^{2-} , Cl^- , Mg^{2+} , Na^+ , Ca^{2+} , Sr^{2+} and the global salinity of water (TDS) were found, suggesting a common origin of evaporite dissolution. Three water groups were successfully identified using PCA and HCA through multivariate statistical analysis; 1) saltwater (23%), where the $TDS > 1000\ mg\cdot dm^{-3}$ with a high concentration of SO_4^{2-} , 2) moderately saline waters also present (51%) but with high concentrations of HCO_3^- , 3) the bulk of the waters are moderately saline (26%) but with a mixed facies.

The use of ionic ratios with various binary diagrams further suggests that the geochemical processes controlling the Remila aquifer system are; evaporite dissolution and/or precipitation (calcite, gypsum, halite, and dolomite) and shall be accompanied by ionic exchange and/or reverse ionic exchange. In the northeastern part of the area, close to Sabkha, an intrusion of

saline waters occurs, which was favoured by intense exploitation of the aquifer by farmers.

The results of this work can be used as a conceptual platform for future research, and to prepare a sustainable development plan in the region taking into account the risks caused by human activity. For example, groundwater over-exploitation can be managed by installing a suitable and economical irrigation system and the planting of drought-tolerant species with the incorporation of unconventional water into the irrigation system.

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