

Groundwater hydrochemical characterization and quality assessment in the Ghriss Plain basin, northwest Algeria

S. Bekkoussa, B. Bekkoussa, J.-D. Taupin, N. Patris and M. Meddi

ABSTRACT

A base line study involving analysis of groundwater samples from the Ghriss Plain Plio-quaternary aquifer, in northwest Algeria, was carried out to determine the major elements chemistry and to assess the hydrogeochemical processes and water quality for domestic use. The main groundwater facies in the area belong to the Mg–HCO₃ and Mg–Cl types. Based on TDS classification, 44.5% of the analyzed samples belong to fresh water and the remaining 55.5% to brackish water categories.

A comparison of groundwater quality parameters in relation to WHO limits for drinking water shows that the concentrations of TDS, Cl⁻ and NO₃⁻ exceed the desirable limits in many water samples. The principal component analysis (PCA) technique reveals the contribution of Cl⁻, Na⁺, Ca²⁺, K⁺ and Mg²⁺ in the salinization process and underlines the contamination of the groundwater with NO₃⁻ in relation to the widespread application of nitrate fertilizers and/or infiltration of untreated urban wastewater. The groundwater quality index (GWQI) shows that 70.5% of the water samples fall within the excellent-to-good category, whereas the remaining samples fall in the poor-to-unsuitable for drinking category.

Key words | Ghriss Plio-quaternary aquifer, groundwater quality, major ion hydrochemistry, multivariate analysis

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INTRODUCTION

Knowledge of groundwater recharge origin, hydrodynamics and geochemical evolution has become an important issue to safeguard the sustainable use of groundwater resources. This is especially true for semi-arid zones and particularly in Mediterranean south-shore countries which are characterized by large water demand, caused by continuous growth in population, intensive agricultural practices and industrial development. Intensive water exploitation conducted to satisfy demand, particularly for domestic and agricultural uses, may result in quality degradation and decreased availability of groundwater due to a scarcity of surface resources which are in many cases not perennial (Liu *et al.* 2015).

Intensive pumping of groundwater leads to the decline of groundwater levels, and may cause excessive salinization

of water resources as observed in recent years (Trabelsi *et al.* 2007). In addition, natural causes (low aquifer recharge due to low and variable precipitation in relation to climate change) and anthropogenic factors (wide fertilizer use caused by intensive agricultural practices, intensive traditional harvesting, extension of urban areas, wastewater use in irrigation, mining, etc.), have caused significant water quality degradation in the last few decades (Kass *et al.* 2005; Caraballo *et al.* 2016). Another source of salinization in aquifers is related to the geological formations, especially when evaporites are present (Ben Ammar *et al.* 2016).

To guarantee integrated water resource management, it is crucial to assess the chemical composition of the water and to control parameters responsible for deterioration of its quality. Classical tools such as chemical composition analysis

(e.g. Piper diagram, Stiff diagram) and statistical approaches such as principal component analysis (PCA) or Q-mode cluster analysis, can reveal component distribution in local water, its spatial chemical evolution and factors controlling the mineralization. Another tool for water quality control for human consumption is the groundwater quality index (GWQI), which is one of the most effective ways to describe the quality of groundwater. This index was developed to summarize water quality data in an easily expressible and understandable format, and was especially developed for groundwater (Zaidi *et al.* 2016; Singh & Hussian 2017).

The study area (Ghriss Plain) is the most productive agricultural zone in the north-western part of Algeria where groundwater is the most widely used resource for agricultural activities and water consumption. However, the quality of this resource has been deteriorating rapidly in the last few years and excessive concentrations of nitrates, chlorides and salinity have been recorded. Anthropogenic factors such as intensive use of fertilizers and untreated wastewater are responsible for the major pollution observed in the region. Consequently, domestic water originating from groundwater shows chemical parameters exceeding standards fixed by international or national agencies, which may cause public health issues. The aim of the present study is to assess the suitability of groundwater for drinking purposes in the Ghriss Plain basin based on computed water quality index values. The study involved comprehension of groundwater mineralization using simple hydrochemical tools and multivariate statistical analysis.

STUDY AREA DESCRIPTION

Localization and climate data

The Ghriss aquifer system is located in the north-west of Algeria (Figure 1). The Ghriss Plain occupies an area of 605 km². It is part of the watershed of Oued Fekan which extends over an area of 1,185 km². The plain corresponds to a flat expanse (470 m average altitude), dominated by high edge reliefs, up to 1,100 m altitude in the south (Nesmoth mountain).

The Ghriss Plain has a semi-arid climate with hot and dry summers, and mild and wet winters. The average

annual temperature is 17 °C. Monthly temperatures range from 23 to 27 °C during the months of June to September, and from 8.8 to 18.6 °C from October to May. The humidity recorded at the Mascara station during the period 1985–2005 varies on average from 46% in July to 75% in December. Rainfall measurements are available from 13 pluviometers, taken between 1976 and 2001. The average annual rainfall for this period is about 313 mm (Bekkoussa *et al.* 2013). Rainfall mainly occurs in the wet season (October–March) and is generally correlated with altitude. The potential evaporation far exceeds the annual rainfall with an annual mean of approximately 877 mm estimated from 1985 to 2004.

The analysis of the rainfall evolution in the study area shows a clear decrease since 1973 which is estimated at 25%. The pluviometric deficit was recurrent over several years, resulting in the reduced flow of the principal river of the Ghriss Plain, the Fekan Wadi, before it totally dried out in the last few years. The water table of the Ghriss Plio-quadernary aquifer decreased drastically by up to more than 60 meters between 1970 and 2010 in some sectors, which can be attributed to both climate change and overexploitation. Indeed, the estimated recharge of the Ghriss Plain aquifer system is 65.3 million m³ per year and the total withdrawn volume is 64.7 million m³ per year (Bekkoussa *et al.* 2008).

The most important economic activity of the area is agriculture; the chief crops are cereals and vegetables. The aquifer is characterized by intensive exploitation and by a great variety of natural and anthropic factors affecting groundwater chemistry.

Geological and hydrogeological context

The geology of the aquifer has been described in detail by Sourisseau (1972) and Assens *et al.* (1977). Ghriss Plain is a hydrogeological entity flanked by reliefs with various geological aspects (Figure 1).

To the north and northwest of the plain lie the southern slopes of the Beni Chougrane mountains composed of very crinkled lands with Cretaceous bones and a thick tertiary recovery. The Cretaceous is represented by marly formations and calcareous marl. It is overlaid by the Miocene formation. The northern and western limits of the Ghriss Plain are

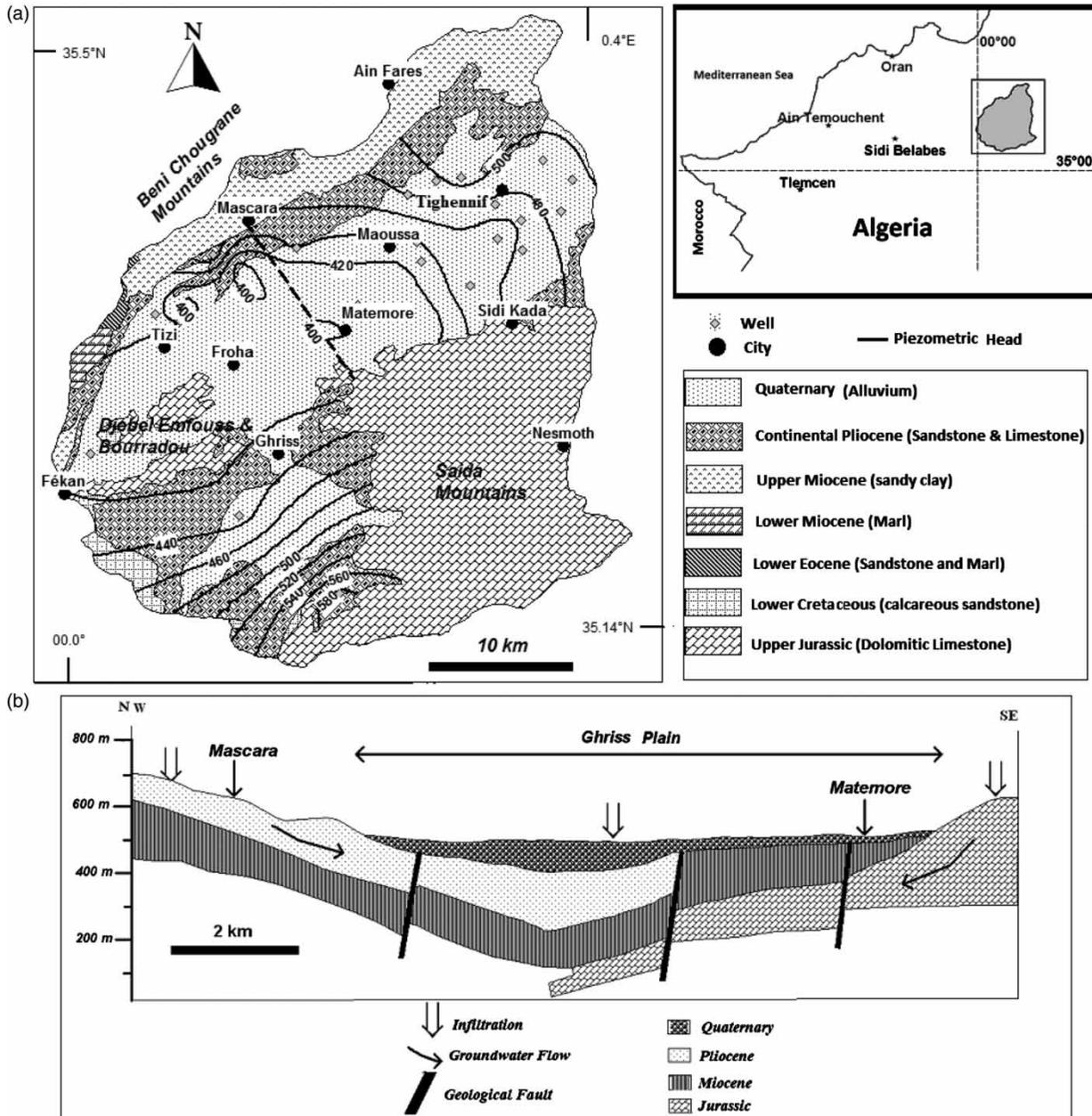


Figure 1 | (a) Geological and piezometric map of the study area and groundwater sample locations. (b) Geological cross section.

formed by marl with some evaporite (gypsum and halite) and argillaceous structures from Miocene, covered by permeable lands contributing to the recharge of the plain aquifers.

In the north-east of the plain, and specifically to the east of the city of Tighennif, the raising of marl and clay formations of the Oligocene and Miocene forms a relatively impermeable barrier which significantly demarcates the

catchment area of the Ghriiss Plain from that of Oued Mina. The southern edge of the Ghriiss Plain is formed by the Saida Mountains. They consist of Jurassic limestone formations that have undergone an intense tectonic shift (horst of Jebel Emfouss and Bourradou, see Figure 1). This set sinks slightly to the north, forming a karst groundwater flow towards the Ghriiss Plain.

The center of the plain is a collapse basin where there are deposits of limestone (especially in the north part of the basin), red clay, fine sands and Quaternary age gravel.

The Ghriss Plain is composed of three superposed aquifers: Plio-quaternary aquifer; the Pliocene limestone and sandstone aquifer; and the Jurassic calcareous dolomitic aquifer.

The Plio-quaternary aquifer, the object of this study, is an unconfined reservoir which extends across the plain. It consists of quaternary alluvium, porous white limestone, sand, sandstone and lacustrine limestone. Natural recharge occurs directly as infiltration from precipitation. A significant lateral flow comes from the Jurassic aquifer in the south-eastern zone and from the Pliocene aquifer in the north. Another form of recharge is excessive irrigation, which is not evapotranspired. The water table depth ranges from -6 m in the north-east to -80 m in the center of the plain and the thickness of the reservoir ranges from 20 m to 150 m (see cross section in Figure 1). This aquifer is bounded by Miocene marls. The direction of groundwater flows converge from north-east and south to south-west zones towards a common outlet on the western boundary called Ain Fekan, a spring which dried in the beginning of 1980's. The rainfall infiltration rate is estimated at about 20%. Transmissivity values range from 5.10^{-5} to 10^{-2} m²/s (Sourisseau 1972). The annual pumping flux from the Ghriss aquifer system decreased from 73.5 million m³ in 1972 to 65 million m³ in 2001 through 3,128 pumping wells and boreholes, due to pumping restrictions. In 1972, the annual recharge of the aquifer system was estimated at 89 million m³ per year; now this recharge has decreased to 65.3 million m³ per year.

The piezometric maps established after a groundwater level survey conducted in September 2010 (Figure 1) indicate a recharge area located in the southern and eastern areas, and a high groundwater discharge in the center (piezometric depression) due to high pumping. The mean hydraulic gradient of the aquifer ranges from 0.75% in the northern sector to 2.5% in the central part of the basin between Matemore and Maoussa cities. A hydraulic gradient of 3.5% is observed in the Guerdjoum region. This important value is explained by a significant decrease in permeability in this zone.

MATERIAL AND METHODS

This study is based on 34 groundwater samplings of the Plio-quaternary aquifer that were collected during July 2008 from farm and public supply wells across the Ghriss basin, and analyzed for major chemical constituents (Figure 1). These samples were taken by ANRH (National Agency of Hydraulic Resources) officers and analyzed by the laboratory of the same organization. Sample bottles were made from high-density polypropylene (HDPP) and were cleaned by rinsing them with distilled water. The water samples were collected after pumping for 10 to 15 minutes in order to remove stagnant groundwater. The pH, electrical conductivity (EC) and total dissolved solids (TDS) were measured *in situ* using pH and EC meters according to standard procedures (Nielsen & Nielsen 2006). The anions (Cl^- , SO_4^{2-} and NO_3^-) were analyzed by ion liquid chromatography and bicarbonate (HCO_3^-) was determined by titration with HCl. The cations (Ca^{2+} , Mg^{2+} , Na^+ , K^+) were analyzed by atomic absorption spectrometry. Careful quality controls were undertaken to obtain a reliable analytical dataset with an ionic balance error within a standard limit of $\pm 5\%$ (Domenico & Schwartz 1998), consequently all 34 samples were used in this study.

The Piper diagram and PCA statistic methods were used to show chemical water characteristics and parameters controlling mineralization processes. PCA is a multivariate statistical method applied to the hydrochemical data of all samples. It is a data transformation technique that attempts to reveal a simple underlying structure that is assumed to exist within a multivariate dataset (Ahada & Suthar 2017; Hamdi et al. 2018).

The GWQI was used to define the suitability of groundwater for drinking purposes. The GWQI method reflects the composite influence of the physicochemical water parameters; it is a dimensionless number and expresses the overall water quality in a given area by combining different water quality variables in a single number. The groundwater quality was measured using the following equation for GWQI with respect to WHO standards (Krishan et al. 2016; Bouderbala 2017),

$$GWQI = \sum SI_i = \sum W_i q_i = \sum \left[\left(\frac{w_i}{\sum_{i=1}^n w_i} \right) * \left(\frac{C_i}{S_i} * 100 \right) \right]$$

Table 1 | Weights of parameters used in GWQI assessment (Zaidi et al. 2016)

Chemical parameters	Weight	Relative weight
pH	3	0.094
TDS	5	0.156
Ca ²⁺	2	0.063
Mg ²⁺	2	0.063
Na ⁺	3	0.094
K ⁺	3	0.094
Cl ⁻	4	0.125
HCO ₃ ⁻	1	0.031
SO ₄ ²⁻	4	0.125
NO ₃ ⁻	5	0.156
Total	32	1

where C_i is the concentration of each parameter, S_i is the limit value admitted according to the standard, w_i is the assigned weight according to its relative importance in the overall quality of water for drinking purposes (Table 1), q_i is the water quality rating, W_i is the relative weight, and SI_i is the sub index of the i th parameter.

The permissible limits for drinking water assigned by the WHO standards have been used for calculating the GWQI in the present study. 10 parameters (pH, TDS, Ca²⁺, Mg²⁺, Na⁺, K⁺, SO₄²⁻, Cl⁻, HCO₃⁻ and NO₃⁻) have been selected. In the first step the 10 selected water quality parameters were assigned a weight on a scale of 5. The assignment of the

weight to a given parameter (Table 1) is set and depends on the relative importance of that parameter influencing the overall drinking water quality and also its impact on human health (Jasmin & Mallikarjuna 2014; Kumar et al. 2014; Bouderbala 2017). The second step involves the assignment of relative weights to each of the parameters. The third step involves the calculation of the quality rating scale (q_i) for each parameter and for each analyzed sample. In the fourth step the sub-index of the i th parameter SI_i is calculated. Finally, in the fifth step the GWQI of the individual water samples is calculated by summing the SI_i values of all the parameters.

RESULTS AND DISCUSSION

Water quality for drinking purposes

General characteristics of groundwater physicochemical parameters for the study area are summarized in Table 2. All of the parameters exhibit a high dispersion, implying a strong spatial variability, as indicated by the high standard deviation values, which is attributed to the presence of fresh, brackish and polluted waters among the groundwater samples.

All the samples ($n = 34$) showed pH values ranging from 6.90 to 8.59 with a mean value of 7.90 ± 0.53 , indicating that the groundwater of the study area is mainly alkaline. For human consumption, water pH is optimum in the range of

Table 2 | Statistics of physico-chemical parameters, major ions ($n = 34$), World Health Organization (WHO 2004) and Algerian Standards (Journal Officiel 2011) for drinking waters of study area

Variable	Minimum	Maximum	Mean	Standard deviation	WHO standards	Algerian standards
Ca ²⁺	28.0	733.0	126.0	152.8	200	200
Mg ²⁺	31.0	407.0	96.7	82.3	150	150
Na ⁺	23.0	713.0	180.6	165.4	200	200
K ⁺	1.0	36.0	6.6	5.9	12	20
Cl ⁻	48.0	3,340.0	518.3	674.1	250	500
SO ₄ ²⁻	47.0	482.0	150.6	94.6	250	400
HCO ₃ ⁻	79.0	323.0	216.3	68.5	–	–
NO ₃ ⁻	4.0	99.0	33.3	27.7	50	50
pH	6.90	8.59	7.90	0.53	6.5–9.2	<9.0
EC (μS/cm)	500	8,630	1,938	1,758	1,500	2,800
TDS	440	7,060	1,539	1,427	1,000	2,000

Ionic concentrations are given in mg/L.

6.5–9.2 according to WHO (2004). All pH values fall within the range admitted for drinking water by both the WHO and the Algerian standards (Table 2).

The EC varied widely and ranged between 500 and 8,630 $\mu\text{S}/\text{cm}$ at 25 °C with a mean of 1,938 $\mu\text{S}/\text{cm}$. More than 44% and 20% of the samples exceeded the maximum conductivity admitted by WHO and Algerian standards for drinking water, respectively.

TDS is an important parameter in drinking water and other water quality standards. TDS in the groundwater of the area ranged from 440 to 7,060 mg/L with an average value of 1,539 mg/L. Freeze & Cherry (1979) classify water into fresh (TDS < 1,000 mg/L), brackish (> 1,000 mg/L), saline (> 10,000 mg/L) and brine (100,000 mg/L) categories on the basis of TDS concentrations. Based on this classification, 44.5% of the groundwater of the study area belongs to fresh water and the remaining 55.5% to brackish water categories.

The nitrate concentration in groundwater and surface water is normally low but can reach high levels as a result of leaching or runoff from agricultural land or contamination from human or animal wastes as a consequence of the oxidation of ammonia and similar sources (Elisante & Muzuka 2016). The nitrate values ranged from 4 to 99 mg/L. Nitrate concentrations for 11 wells (32% of samples) exceeded the maximum permissible value of 50 mg/L indicated by the WHO and the Algerian standards. The agricultural activities are the likely source responsible for high concentrations of nitrates in the study area.

Chlorine is the element that most affects the groundwater quality in the study area. Indeed, we recorded 20 wells (59%) with a concentration of Cl^- ions, exceeding the WHO water standards. The Algerian standard regarding Cl^- is more flexible and the number of wells above the maximum concentration is 8 samples.

Taking into account mean concentrations across the study zone, Na^+ was the dominant cation (180.6 mg/L), followed by Ca^{2+} (126.0 mg/L), Mg^{2+} (96.7 mg/L) and K^+ (6.6 mg/L). Cl^- was the dominant anion (518.3 mg/L) followed by HCO_3^- (216.3 mg/L) and SO_4^{2-} (150.6 mg/L).

Chemical water types

From the Piper diagram (Figure 2), various hydrochemical facies were identified. Mg- HCO_3 and Mg-Cl are the

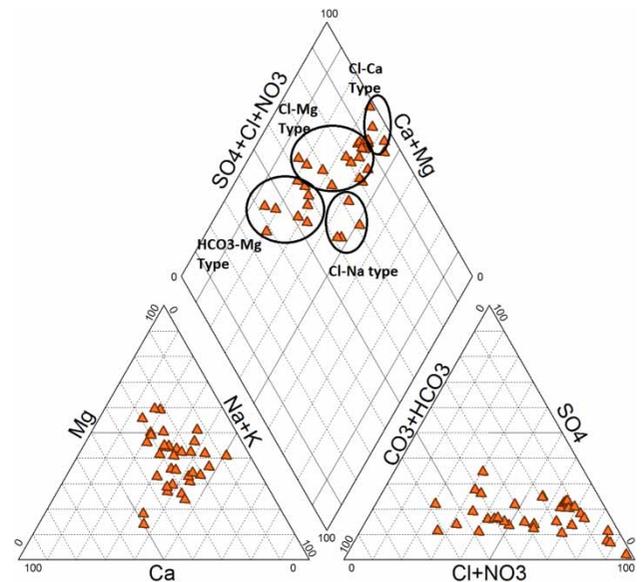


Figure 2 | Groundwater facies classification using Piper plot.

dominant types of water, each of them representing 26% of all samples, while Na-Cl and Ca-Cl types represent 13% and 3%, respectively. The Mg- HCO_3 facies characterize lateral recharge and vertical leakage from Jurassic carbonate formations. It was observed on the southern edge of the Saida Mountains, near the Emfous and Bouradou horsts (Figure 1) and in some wells situated in the center of the plain. This facies is typically representative of the karst waters origin. It is characterized by lower EC and TDS and higher concentration of HCO_3^- .

Mg-Cl type is a mixture of waters from a dolomitic environment (hence the predominance of magnesium) as well as seepage from effective rainfall and/or return of irrigation water. Indeed, the origin of the dominant anion (Cl^-) in this type of water is in all likelihood superficial (anthropogenic contamination or evaporitic contamination by halite originating from the Miocene formation), while the ubiquity of magnesium can only be explained by a recharge coming from the aquifer of dolomitic limestone of the Jurassic period.

The Ca-Cl facies was mainly found in the northeastern part and south of the plain. These waters are highly mineralized and salinity reached more than 8 g/L in one well. This facies is representative of groundwater characterized by an anthropogenic pollution with high concentrations of chlorides, potassium and nitrates.

Na-Cl-type water is mainly localized in the east and center of the plain. The evaporites formation dissolution, especially halite, and cation exchange reactions clearly influence the hydrochemical composition of this type of water in different proportions. In fact, the Na-Cl ratio in the majority of the samples for this type of water is between 0.8 and 1.0, which is close to the theoretical dissolution line of halite (Bouderbala & Gharbi 2017).

Principal component analysis (PCA)

To accurately identify the main processes governing the hydrochemistry of groundwater in the system, a factor analysis was performed using a subset of nine variables, namely the major ion concentrations (Ca^{2+} , Na^+ , K^+ , Mg^{2+} , Cl^- , SO_4^{2-} , HCO_3^- and NO_3^-) and total mineralization (TDS).

PCA was performed for the 34 samples of the Ghriss Plain Plio-quaternary aquifer. The first two factors explain 78.83% of the total dataset variance (Table 3). The number of principal components kept was based on the Kaiser criterion, for which only the components with eigenvalues greater than 1 are retained (Kaiser 1958).

F1 accounts for 61.61% of the variance, and is strongly positively loaded with TDS (0.98), Cl^- (0.98), Na^+ (0.96), Ca^{2+} (0.90), K^+ (0.89) and Mg^{2+} (0.85). This clustering

of variables associated with F1 suggests that salinization may be the major process controlling the hydrochemical variability, and connotes salinity enhancement most likely arising from mineral dissolution. It indicates also the important contribution of these ions to the mineralization of groundwater through the water-rock interaction processes.

The factor F2 explains 13.6% of the total variance, showing a significant positive association with HCO_3^- (0.86), which provides insight into the importance of the recharge coming from the karstic aquifer. The second factor is found to be strongly negatively loaded with nitrate NO_3^- (-0.72), which represents contamination from domestic waste and agricultural activities. Higher concentrations of nitrogen compounds are usually an indication of intensive agricultural use (e.g. arable farming and vegetable growing) and extensive urbanization with improper sewage systems. This result reveals two competing types of recharge influencing the hydrochemistry of the groundwater: superficial recharge with effective rainfall and excess irrigation water marked by nitrate, or lateral recharge and vertical leakage from carbonate formations of the Jurassic period marked by HCO_3^- . The omnipresence of NO_3^- in the PCA analysis emphasizes anthropogenic processes in relation to the widespread application of nitrate fertilizers and/or infiltration of untreated urban wastewater.

Estimation of groundwater quality index (GWQI)

The GWQI values ranged from 36.4 to 415.4 with a mean value of 100.1. Results reveal that about 26.5% of samples report excellent water quality type (GWQI < 50), and 44.1% present good water quality type (GWQI = 50–100), whereas 17.6% exhibit poor water quality (GWQI = 100–200), 8.8% of the water show very poor quality type (GWQI = 200–300), and the remaining 3% indicate water unsuitable for drinking purposes (GWQI > 300).

Excellent water quality type samples are located in the central and southern part of the Ghriss Plain (Figure 3). However, good quality samples are distributed in different parts of the studied area. Poor, very poor and unsuitable water quality samples are all localized in the north-west and in the east, which was predictable, given the high salinity of the water in these zones.

Table 3 | Principal component analysis for groundwater samples

	F1	F2
Ca^{2+}	0.909	-0.125
Mg^{2+}	0.858	0.391
Na^+	0.964	0.158
K^+	0.896	0.003
Cl^-	0.984	0.072
SO_4^{2-}	0.480	0.422
HCO_3^-	-0.292	0.864
NO_3^-	0.432	-0.725
pH	-0.651	0.260
TDS	0.986	0.085
Eigenvalues	6.161	1.723
% of variance	61.609	17.226
Cumulative %	61.609	78.835

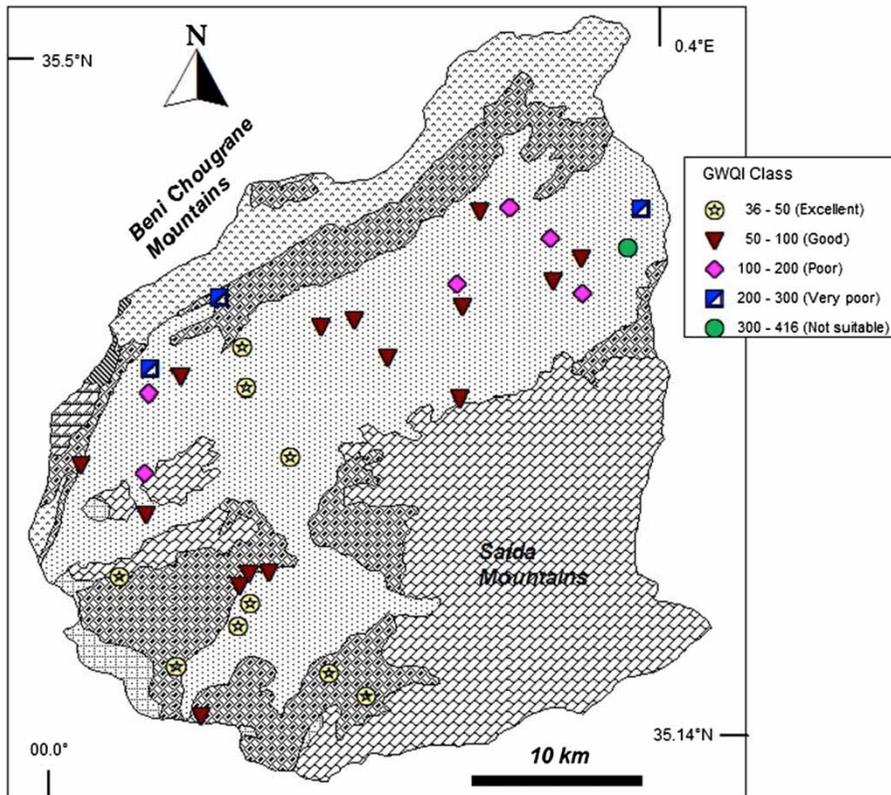


Figure 3 | GWQI classes map.

The correlation between the physico-chemical parameters and the GWQI index shows that TDS, Cl^- , Na^+ and Ca^{2+} are strongly correlated ($R > 0.9$) to GWQI indicating the impact of these parameters on the quality of water in this region. K^+ and Mg^{2+} show moderately strong correlation (R between 0.8 and 0.9). GWQI shows a correlation of less than 0.5 for parameters like SO_4^{2-} and NO_3^- . However, HCO_3^- and pH show an inverse correlation with GWQI. This can be explained by the fact that the recharge flow coming from the carbonate layer, marked by relatively high concentrations of HCO_3^- and basic pH, has a good quality which positively influences the overall quality of the studied aquifer.

CONCLUSION

The main conclusions from this study can be summarized as follows.

Groundwater hydrochemistry in the studied aquifer is dominated by the Mg- HCO_3 and Mg-Cl water types. Cl^- is

the element that most affects the groundwater quality in the study area.

The application of multivariate analysis (PCA) shows that anthropogenic (surface runoff, agriculture fertilizers) and natural/geogenic sources (water-rock interactions) are the main processes responsible for variations of physico-chemical parameters in this aquifer.

According to the GWQI assessment, the majority of the samples rated are good to excellent water quality types, whereas very poor to poor water quality for drinking purposes were principally situated in the north-east.

A thorough investigation based on geochemical and modeling tools must be done to illustrate mechanisms and factors impacting total mineralization of water in the studied aquifer, especially in polluted and/or vulnerable zones. Minor and trace elements can also be taken into account in future groundwater quality studies. These results could be used to define specific actions for the adequate management, protection and, if necessary, restoration of the associated water reservoirs.

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