

Hydrogeological and mixing process of waters in deep aquifers in arid regions: south east Tunisia

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Abstract This study addresses the hydrogeochemistry of thermal and cold waters from south east Tunisia. Temperature intervals are 38.5–68 °C and 22–27.8 °C for thermal water and cold water, respectively. Three distinct hydrogeological systems supply water either for irrigation or for drinking; they are: (1) the Continental Intercalaire geothermal aquifer (CI), (2) the Turonian aquifer and (3) the Senonian aquifer. A synthetic study including hydrochemical, hydrogeological and geothermal approaches have been applied in order to evaluate the inter-aquifers water transfer in south east of Tunisia. By using silica geothermometers and saturation indices for different solid phases, estimated thermal reservoir temperature varies between 52 and 87 °C and between 75 and 110 °C, respectively. Based on chemical and thermal data, mixing, which occurs between the ascending deep geothermal water and shallow cold water, is about 57 % cold water.

Keywords Thermal water · Mixing process · Geothermometry · Saturation indices · Tunisia

Introduction

Geothermal well waters are sometimes the result of fluid mixing in the reservoir and/or in the well (Truesdell 1975). In these cases, it is impossible to obtain numerically a unique solution to the problem of origin, in terms of chemical characteristics, estimated temperature in depth... etc., without assuming that all of the given chemical components are of the same origin.

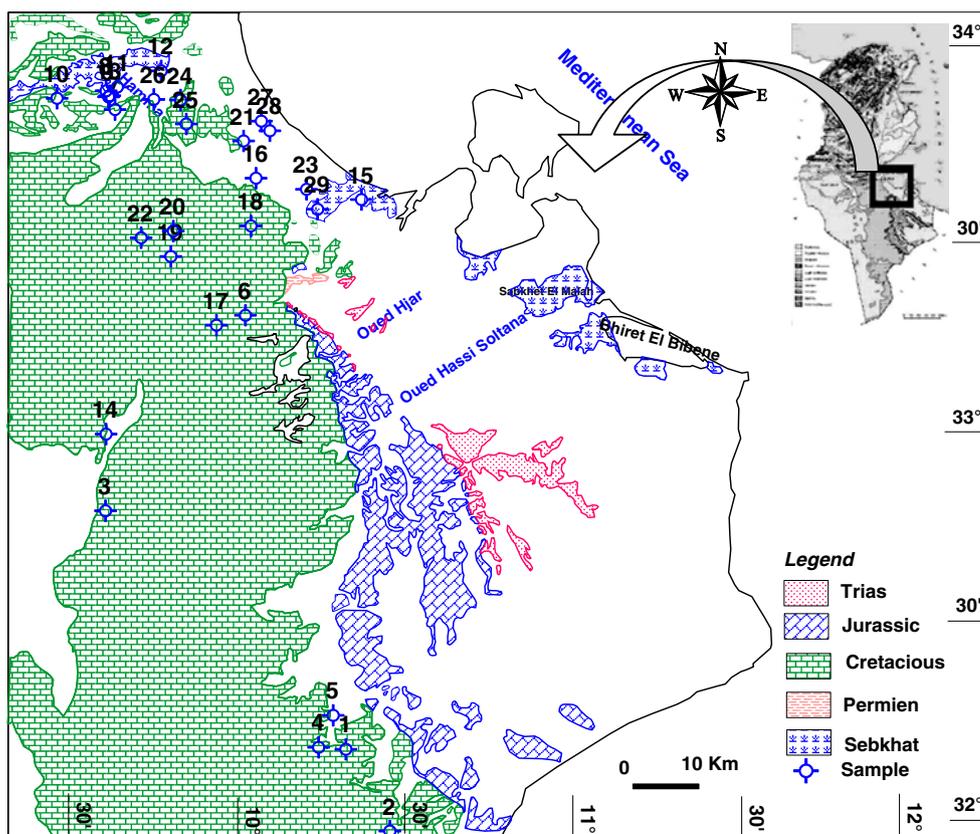
The mixing between shallow and thermal groundwater and gain or loss of steam, may modify the chemistry of the ascending fluids and lead to erroneous interpretations (Fournier and Truesdell 1974; Amorsson 1983). Several workers have proposed a method that uses the chemical composition of the water to find the ratio of the fraction mixing and the origin temperature (e.g. Fournier and Truesdell 1974; Reed and Spycher 1984). In addition, the chemistry of thermal waters has attracted the attention of numerous studies, in particular investigations of the influence of water–rock interactions and the large diversity of the ionic composition of fluids that are found in geothermal systems (e.g. Mahon 1970; Tonani 1970; White 1970; Fournier and Truesdell 1973; Ellis and Mahon 1977; Fournier and Potter 1979; Giggenbach et al. 1983; Giggenbach 1988, Pauwels et al. 1993; Tarcan and Gemici 2003).

Tunisia has an important thermal potential and is well-known since the Roman period. Tunisia is known with more than 90 thermal manifestations including 50 thermal springs. South east Tunisia constitutes an important hydrothermal basin. This region was the object of different hydrogeothermic studies (Makni 2006; Bouri et al. 2008; Makni et al. 2012). The most important aquifers hosted in Cretaceous formations made up of sandstone (Continental Intercalaire aquifer) and dolomite limestone (Turonian and Senonian aquifers) (Ben Baccar 1982). Continental Intercalaire (CI) aquifer is characterised by thermal water, whereas Turonian and Senonian aquifers supply cold waters.

To assess hydrogeothermal resources of Southern east Tunisia, hydrogeothermal study comprising geological, hydrogeological, hydrogeochemical and geothermometric approaches have been realized. This approach is important for the determination of the different hydrogeochemical facies of thermal and cold waters, and to relate them to their origin reservoirs. It provides a good tool for the determination of hydrothermal basins.

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Fig. 1 Geological map of the study area



One of the major tasks in the exploration of geothermal resources is the estimation of the subsurface temperature using geochemical composition of thermal springs and the evaluation of the ratio of mixing water.

This paper aims to characterise thermal and cold waters in order to study the eventual water exchange between the three deep aquifers using a hydrogeothermal approach.

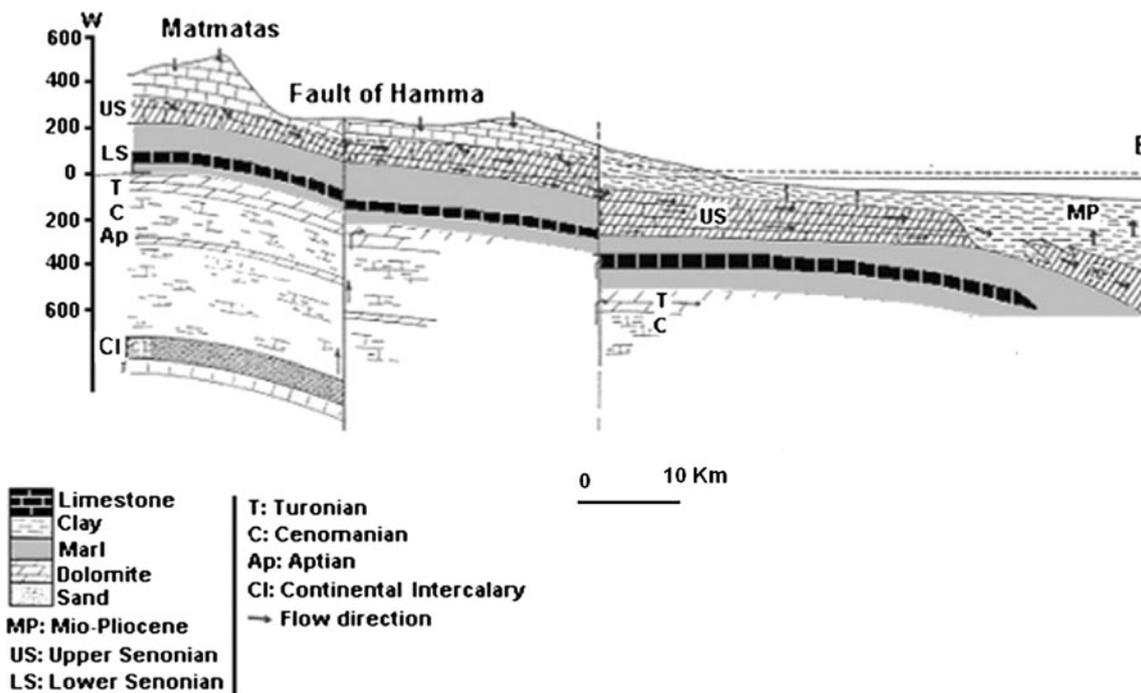


Fig. 2 Schematic geological cross section through the Gabes-south aquifer (UNESCO 1972)

Study area

South east Tunisia (Fig. 1) is characterised by an arid climate with annual average precipitation of 189 mm, occurring mainly during the winter, between October and January. The age of geological formations cropping out in south east Tunisia extends from the upper Permian to the Quaternary.

According to Burollet and Desfoges (1982) and Ben Ayed (1986), the south-eastern Tunisia basin has undergone a complex and polyphase structural history since the Carboniferous era. It has been affected by multiple episodes of tectonism, including a late Paleozoic collision with Laurasia and subsequent early Mesozoic rifting associated with the opening of the Tethyan Ocean (Memmi et al. 1986; Gabtni et al. 2006). Several tectonic phases affected the study area and produced a large sedimentary basin with a fault-bounded structural high (Gabtni et al. 2009). Numerous geologic studies in

south east Tunisia have been important in defining the regional and local geology and the geometries and boundaries of potential reservoirs. The investigated area is located in the north-eastern Saharan Domain where the general tectonic lineaments strike NW–SE with conjugated NE–SW faults that lead to the discharge of hot water in the study area (Bouri et al. 2008).

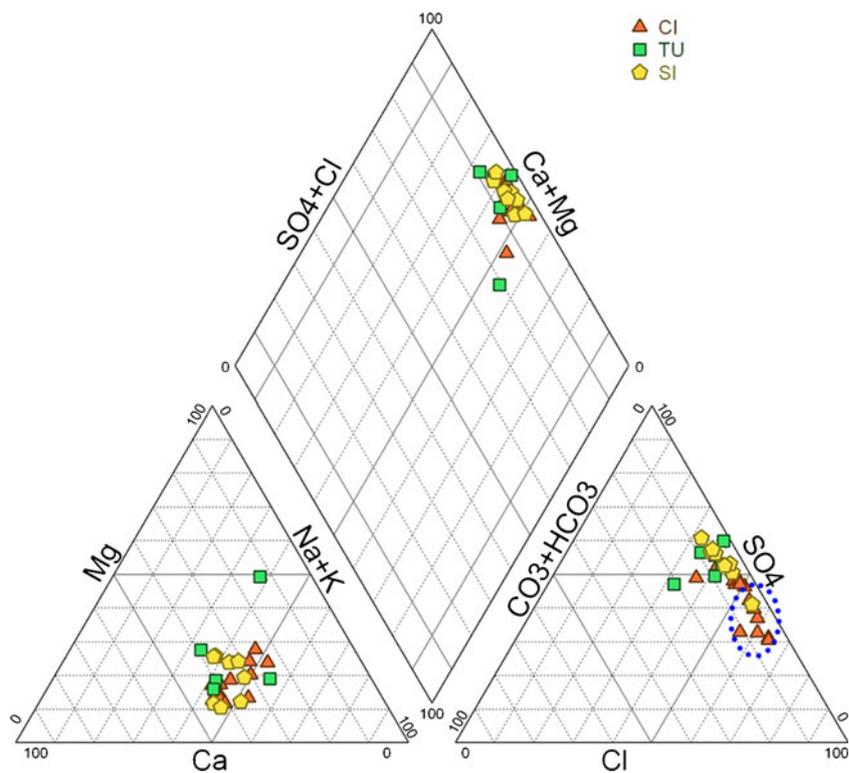
Available data from geological and geophysical studies have provided the means to identify the main potential hydrogeological entities (Fig. 2):

- (a) The Upper Cretaceous, represented by the Cenomanian, the Turonian (TU) and the Lower Senonian (SI). The SI is divided into two distinct entities: the marl-gypsum Senonian and the limestone Senonian. Water temperature supplied from these units varies between 22 and 27.8 °C.
- (b) Lower Cretaceous, mainly, the CI aquifer is formed by sandstone. It is situated within the continental

Table 1 Chemical composition of the south-eastern waters (in milligrams per liter)

Samples no	Aquifer	<i>T</i>	TDS	pH	Ca	Mg	K	Na	HCO ₃	Cl	SO ₄	SiO ₂
1	CI	26	1.73	7.88	245.39	42.79	41.82	258.11	122.00	652.51	477.74	5.3
2	CI	27	1.63	7.5	195	61.4	33	263	171	500	397	3.8
3	CI	32.9	3.252	8.04	336	137	43	612	154	1140	974	5
4	CI	26.6	1.312	7.4	101	61.1	8.4	249	169	255	458	5.2
5	CI	28	2.31	7.3	319	80.3	44.9	339	107	850	559	5.2
6	CI	26.2	2.32	8.05	302.00	81.00	51.00	356.00	117.00	880.00	562.00	3.3
7	CI	63.5	2.981	6.7	338	80	59	564	98	910	1130	14.9
8	CI	58	3.33		391.4	71.9	41.8	491.5	116.1	779.9	1180.9	17.16
9	CI	68	2.702		325.5	66.2	42.3	388.9	122.2	655.8	915	25.3
10	CI	58	3.674		488	80.25	75	550	170.8	923	1233	33
11	CI	44	3.988		500	97	80	585	137.2	1065	1135	33
12	CI	40	3.76		424.65	74.2	99.13	506.57	120	883.83	1187.1	30
13	CI	63	2.024		320	82	51	350	97.6	710	920.36	7.8
14	CI	33.5	5.85		434	222	31.7	807	184	1460	1400	33
15	CI	38.5	2.9		200	133.7	13.26	420.67	170.8	564.5	994	33
16	TU	27	2.622	7.74	301.7	85.2	10.7	357.6	36.6	484.2	1023.5	11.9
17	TU	27.2	0.7		99.4	44.06	15	234.93	225.7	204.04	402.18	7.2
18	TU	22.7	2.04	8.1	257.22	110.14	16	243.12	183	400.54	891.48	8.6
19	TU		1.77	8	172.3	78	14	262	148	385	625	5.83
20	TU		1.22	8	97	222	10	315	131	470	925	4.41
21	SI	26.5	2.77	7.28	359	83.1	36.8	396.5	140	599.2	1042.1	4.47
22	SI	24	3.326	7.95	345.5	148	23.7	404.7	183	497.9	1266.7	4.46
23	SI	26.8	2.605	8.4	212	76.7	26.7	351.2	140.3	451.2	940.7	5.43
24	SI	41.6	3.17	7.72	420.6	68.2	51.5	459.2	116	731.6	1104.7	15
25	SI	26.7	3.53	7.01	339.1	68.2	53.5	512.6	110	759.5	1258.2	10
26	SI	32.5	5.914		686.5	102.2	57.2	834.2	170.8	1443.8	1458.9	5.16
27	SI	23.8	3.832	7.33	275.2	116.5	15.9	387.1	143	554.8	1107.5	5.74
28	SI	24.2	2.88	7.49	249.3	117.6	14.4	401.7	142.4	515.8	1089.5	5.62
29	SI	24	3.74	7.85	385	162	14	443	162	790	1324	5.56

Fig. 3 Piper diagram of the study area groundwater samples



formations of Lower Cretaceous (Neocomian, Barremian, Aptian and Albian). The aquifer is composed by a complex succession of detrital sediments separated by clay-rich strata, with the maximum aquifer thickness exceeding 1,500 m (Edmunds et al. 2003).

Two networks of faults characterise the region of south east Tunisia: those of NW–SE direction (the Medenine fault being the most important) and those of NE–SW direction that are faults of downfall or adjustment. The draining of SI water is done by the CI water by these faults (Mamou 1990).

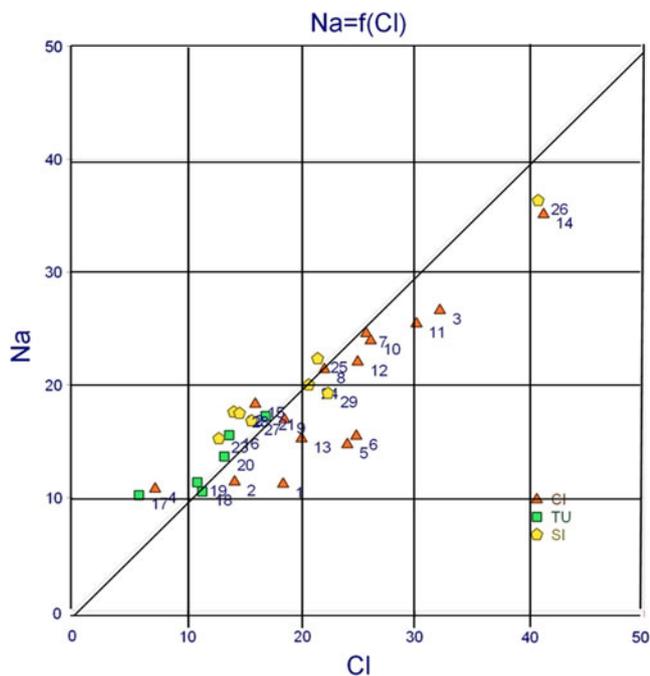


Fig. 4 Relationships between Cl and Na ions in the groundwater samples

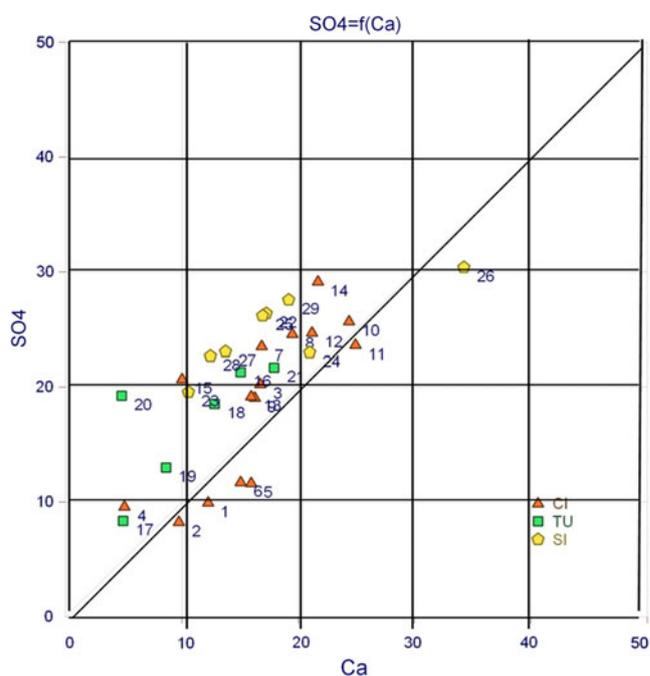
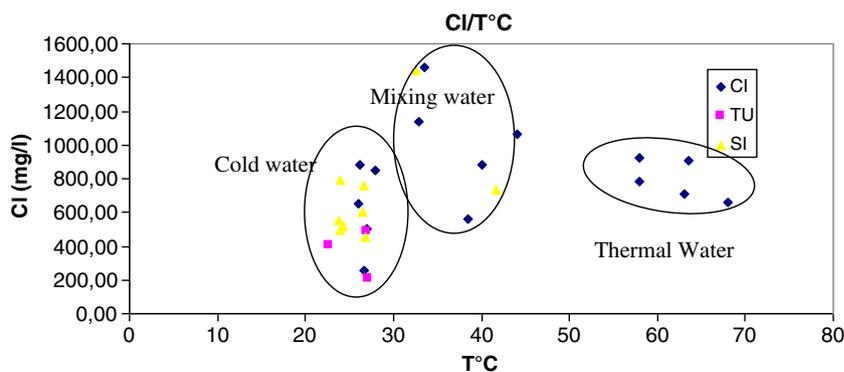


Fig. 5 Relationships between Ca and SO₄ ions in the groundwater samples

Fig. 6 Relationships between temperature and chloride ions in the groundwater samples



Water sampling and analysis

Several analyses such as the water temperature, pH, electrical conductivity (EC), and total alkalinity as HCO₃ were carried out on-site. All samples for chemical analyses were collected in low density polyethylene bottles and filtered in the laboratory through 0.45-μm membrane filters. Samples revealing relatively high salinity (exceeding 3 g/l) were diluted before analysis.

Each sample was collected in two new 500-ml polyethylene bottles. All sampling bottles were washed with de-ionised water and again with filtered sample water before filling it to capacity. For each sample, one bottle is acidified (until pH of samples reached 1) with 35 % nitric acid for cation analysis (Na⁺, K⁺, Ca²⁺ and Mg²⁺), whereas the other is used for the determination of dissolved anions (Cl⁻, SO₄²⁻, HCO₃⁻ and SiO₂⁻). Prior to analysis in the laboratory, the samples were stored at a temperature below 4 °C.

Chloride was determined by the standard titration method or the Mohr method. Bicarbonates were determined by the potentiometric method. Sulphate concentration was measured by the gravimeter method using BaCl₂. Sodium and potassium concentrations were determined with a flame photometer. Calcium and magnesium ions are determined by the complexometric method using ethylene diamine tetraacetic acid bisodium salt. Fluorine concentrations were measured by the calorimetric method. Total dissolved solids (TDS) were measured by evaporating a pre-filtered sample to dryness.

Table 2 The principal component analysis results

N°	Eigenvalue	Cumulative variance
1	58,450	58,450
2	20,379	78,829
3	10,599	89,428
4	5,246	94,674
5	3,113	97,788
6	1,388	99,176
7	0,665	99,841
8	0,159	100,000

Twenty nine samples were collected from thermal and cold wells and spring (Table 1, Fig. 1). The quality of the analyses was evaluated using the ion balance (IB) and only ones with IB<5 % were considered.

Results and discussion

Water chemistry

The chemical composition of the thermal waters is given in Table 1. Total dissolved solids values were 1.7–5.8 g/l. Water temperature ranges from 22 °C, in the selected fresh cold water, to 68 °C, in thermal water. All water samples exhibited a pH between 6 and 8.

The Piper trilinear diagram (Piper 1944) is given in Fig. 3 and it shows that the dominant cations are mainly calcium and sodium, while the dominant anions are sulphate and chloride. The sum of cations and anions shows two main groundwater types: Na–Cl and Ca–SO₄. The Piper diagram

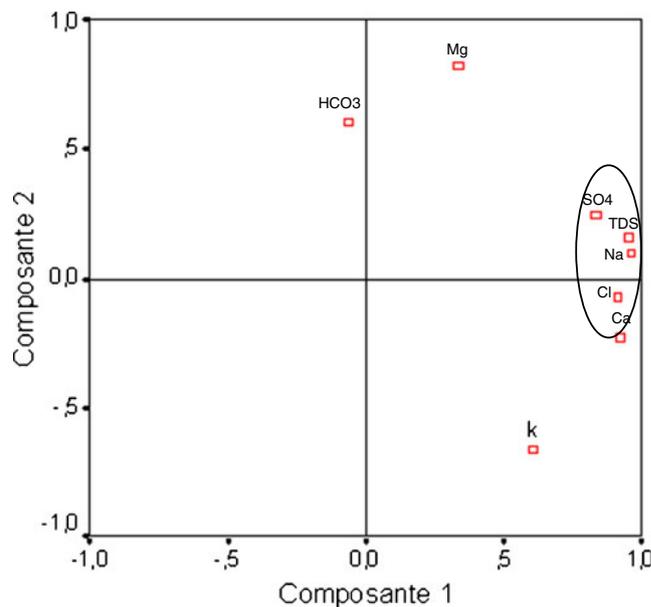


Fig. 7 Projection of variables onto the factorial plane (F1×F2)

Table 3 Correlation matrix 1

Correlation	TDS	Ca	Mg	K	Na	HCO ₃	Cl	SO ₄
TDS	1.000							
Ca	0.853	1.000						
Mg	0.370	0.057	1.000					
K	0.410	0.676	-0.309	1.000				
Na	0.917	0.821	0.365	0.501	1.000			
HCO ₃	0.041	-0.114	0.234	-0.246	0.037	1.000		
Cl	0.822	0.827	0.296	0.615	0.909	-0.068	1.000	
SO ₄	0.844	0.722	0.472	0.291	0.775	-0.070	0.571	1.000

shows that the waters from the Lower Cretaceous aquifer are situated close to the Na–Cl vertex, probably due to the leaching of evaporitic minerals. However, waters from both the wells and Zarat spring (Senonian aquifer) are Na–(Ca)–Cl–(SO₄) mixed type, possibly due to the presence not only of chloride minerals but also of gypsum and/or anhydrite (CaSO₄) in this aquifer. Furthermore, bivariate diagrams of major elements were used in order to separate different mechanisms that contribute to groundwater mineralization. In particular, sodium versus chloride and calcium versus sulphate show positive correlations (Figs. 4 and 5). This indicates the possible dissolution of halite in CI aquifer, gypsum and/or anhydrite-bearing rocks relatively abundant especially in the Senonian and Turonian aquifers.

With the purpose of distinguishing the major groups of water, the correlation between chlorite and temperature (Fig. 6) reveals three groups: cold water (SI and TU on majority), mixing water (between CI and SI) and thermal water (CI).

The multivariate statistical analysis is a quantitative and independent approach of groundwater classification allowing the grouping of groundwater samples and the making of correlations between chemical parameters and groundwater. Correlation coefficient is a commonly used measure to establish the relation between independent and dependent variables (Nair et al. 2005).

Principal component analysis (PCA) was performed on a data set of 29 samples and the eigenvalues and the percentage of the variance explained by each eigenvector is listed in Table 2. Two principal components or factors (eigenvalue) explaining 78.7 % of the variance or information contained in the original data set was retained, which is sufficient to give a good idea of the data structure.

Factor 1 (F1) explains more than 58.4 % of total variance and contains large loadings on Na⁺, Cl⁻, Ca²⁺, SO₄²⁻ and TDS. It represents the weathering of halite and evaporates minerals from the underlying geology. Factor 2 contributes to 20.3 % of the total variance and is strongly associated with K⁺, Mg²⁺ and pH.

The scores of variables and observations onto the two principal component axes are plotted in Fig. 7, which enables identification of several processes of water mineralization.

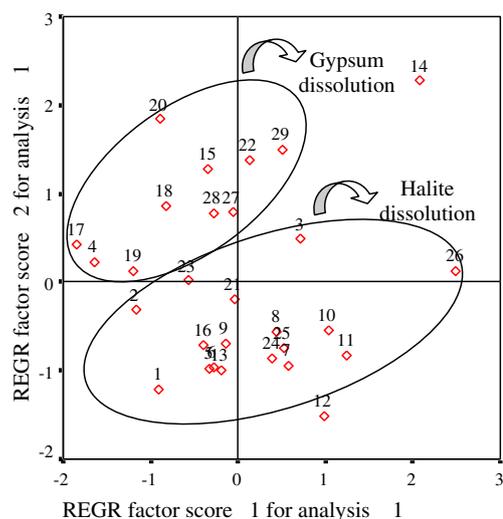
The contents of chloride, sodium, calcium and sulphates are positively correlated to salinity in all waters, with correlation coefficients of 0.82, 0.91, 0.85 and 0.84, respectively. Sodium and chloride are correlated with a coefficient of 0.9 (Table 3). They have a common natural origin that would be mainly the dissolution of the halite. Also, the SO₄ and the Ca have a correlation coefficient of 0.7, indicating that the origin is the dissolution of CaSO₄.

Individual representation (samples) in the first factorial plan (F1, F2) classifies the water in two different groups (Fig. 8):

- A group representing the trend of halite dissolution essentially for CI water;
- A group representing the trend of gypsum dissolution essentially for Senonian and Turonian waters.

Geothermometry

Dissolved chemical species were widely used to obtain information on subsurface temperatures using multiple fluid–mineral equilibrium calculations. Chemical geothermometers depend upon the water–mineral equilibrium and give the last equilibration temperature in the reservoir (Nicholson 1993).

**Fig. 8** Projection of samples onto the factorial plane (F1 × F2)

Several types of chemical geothermometers are described in the literature (e.g. Rybach and Muffler 1981; Kharaka and Mariner 1989; Fournier and Potter 1979).

The correlation matrix of physical parameters (T, pH and TDS) and SiO₂ has been given in Table 4. This PCA reveal a good correlation between T and SiO₂ ($r=0.69$). The projection of variables onto the factorial plan (F1–F2) reveals the good correlation between the temperature and the Silica (Fig. 9). The representation of individual spaces (Fig. 10) reveals three types of water: thermal water (from the CI aquifer), cold water (From Senonian and Turonian aquifers) and mixing water.

One practical and often used application of surface water geochemical analyses is the calculation of subsurface temperatures using chemical geothermometers. Several types of these geothermometers have been studied. According to Fournier (1977), silica is ideal for this purpose as it is generally in excess supply (as it is the main constituent of the vast majority of minerals). He compiled various equations relating temperature with solubility with guidance as to their usage for amorphous silica, quartz (both with and without steam loss), cristobalite (α and β types) and chalcedony.

The good correlation between SiO₂ and temperature can be a good tool to estimate the reservoir temperature of the thermal aquifer. In fact, quartz geothermometers indicate relatively adequate temperatures (Table 5). It reveals an estimated temperature between 30 and 60 °C, for the TU and SI aquifers, and between 52 and 87 °C for the CI aquifer, with a good correlation between the emergency temperatures and the estimated ones (Fig. 11).

Fluid–mineral equilibria

Modelling of the state of equilibrium at different temperatures is another approach to estimate the temperature in depth, especially in the presence of carbonate and evaporate host rocks (D’Amore et al. 1987; Lopez-Chicano et al. 2001).

Equilibrium between an assemblage of minerals constituting an aquifer and a groundwater depends on the temperature. Formulating a numerical model to calculate the saturation state of various likely aquifer minerals at a specified temperature, in a particular groundwater, allows gauging the degree of fluid–mineral equilibrium. By recalculating over a range of temperatures, we can deduce

Table 4 Correlation matrix 2

Correlation	T	TDS	pH	SiO ₂
T	1.000			
TDS	0.210	1.000		
pH	-0.581	-0.120	1.000	
SiO ₂	0.692	0.213	-0.565	1.000

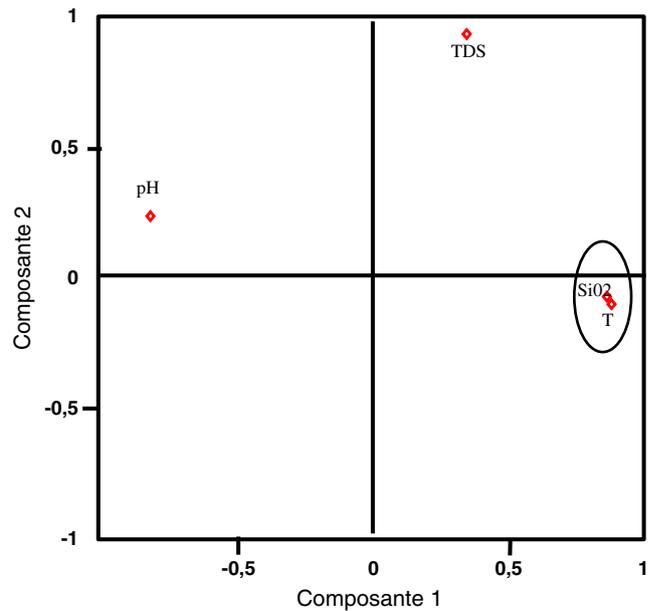


Fig. 9 Projection of variables onto the factorial plane (F1 × F2)

saturation indices ($SI = \log Q/K$; Q the ion activity and K the equilibrium constant of the mineral) as a function of temperature. If the saturation index of a group of minerals converges to zero at a particular temperature, then the temperature most likely corresponds to the equilibration temperature, and the minerals represent the aquifer mineral assemblage (Crerar 1975; Reed 1982; Reed and Spycher 1984; D’Amore et al. 1987; Tole et al. 1993). This approach is called multi-component geothermometry. Pang and Reed (1998) refined the technique further, to overcome some problems associated with erroneous or absent analyses in geothermal waters. This approach represents a modified SI versus temperature graph that eliminates problems with water analyses lacking or with erroneous data.

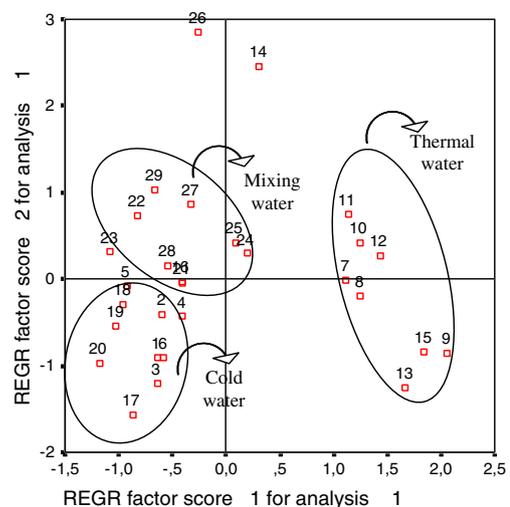
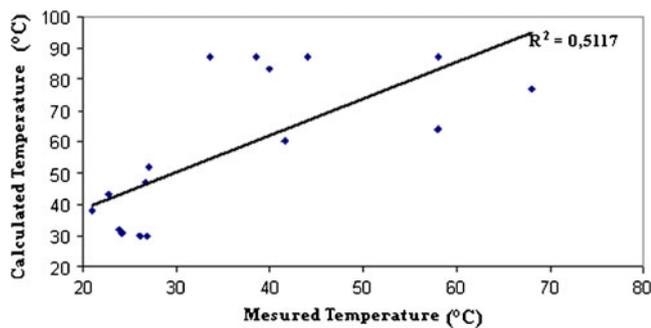


Fig. 10 Projection of samples onto the factorial plane (F1 × F2)

Table 5 Reservoir temperature (°C) estimated using of the Silica geothermometers

Samples	Aquifer	SiO ₂ (mg/L)	T (°C)	QSPV	QAPV	Chalcedony
1	CI	5.3	26	20	30	-13
2	CI	3.8	27	11	21	-22
3	CI	5	32.9	18	28	-15
4	CI	5.2	2.6	19	29	-13
5	CI	5.2	28	19	29	-13
6	CI	3.3	26.2	7	18	-26
7	CI	14.9	63.5	53	59	20
8	CI	17.16	58	58	64	26
9	CI	18.93	68	61	67	29
10	CI	33	58	83	87	52
11	CI	33	44	83	87	52
12	CI	30	40	79	83	48
13	CI	7.8	63	31	40	-1
14	CI	33	33.5	83	87	52
15	CI	33	38.5	83	87	52
16	TU	11.9	27	45	52	12
17	TU	7.2	21	29	38	-4
18	TU	8.6	22.7	34	43	2
19	TU	5.83		23	32	-10
20	TU	4.41		15	25	-18
21	SI	4.47	26.5	15	25	-18
22	SI	4.46	24	15	25	-18
23	SI	5.43	26.8	21	30	-12
24	SI	15	41.6	53	60	21
25	SI	10	26.7	39	47	7
26	SI	5.16	32.5	19	29	-14
27	SI	5.74	23.8	22	32	-11
28	SI	5.62	24.2	22	31	-11
29	SI	5.56	24	21	31	-12
23	SI	5.43	26.8	21	30	-12
24	SI	15	41.6	53	60	21
25	SI	10	26.7	39	47	7
26	SI	5.16	32.5	19	29	-14
27	SI	5.74	23.8	22	32	-11
28	SI	5.62	24.2	22	31	-11
29	SI	5.56	24	21	31	-12

QSPV quartz-no steam loss,
QAPV quartz-maximum steam
loss at 100 °C

**Fig. 11** Correlation between measured and calculated temperatures

According to the mineral equilibrium approach, mineral saturation curves were established for three thermal waters of south east Tunisia (one spring and two wells) (Fig. 12).

For the Zarat spring, saturation indices of the water with respect to quartz, chalcedony, aragonite, calcite, gypsum, anhydrite and magnesite tend to get closer to zero around the temperatures of 75–100 °C (Fig. 12). For the Borg well, the temperature estimated using the multiple mineral equilibrium approach is about 75–100 °C by the magnesite, anhydrite, quartz and the chalcedony minerals. Therefore, we conclude that temperatures of about 75–100 °C represent the temperature of the waters in the deep geothermal reservoir for the Sghayer well.

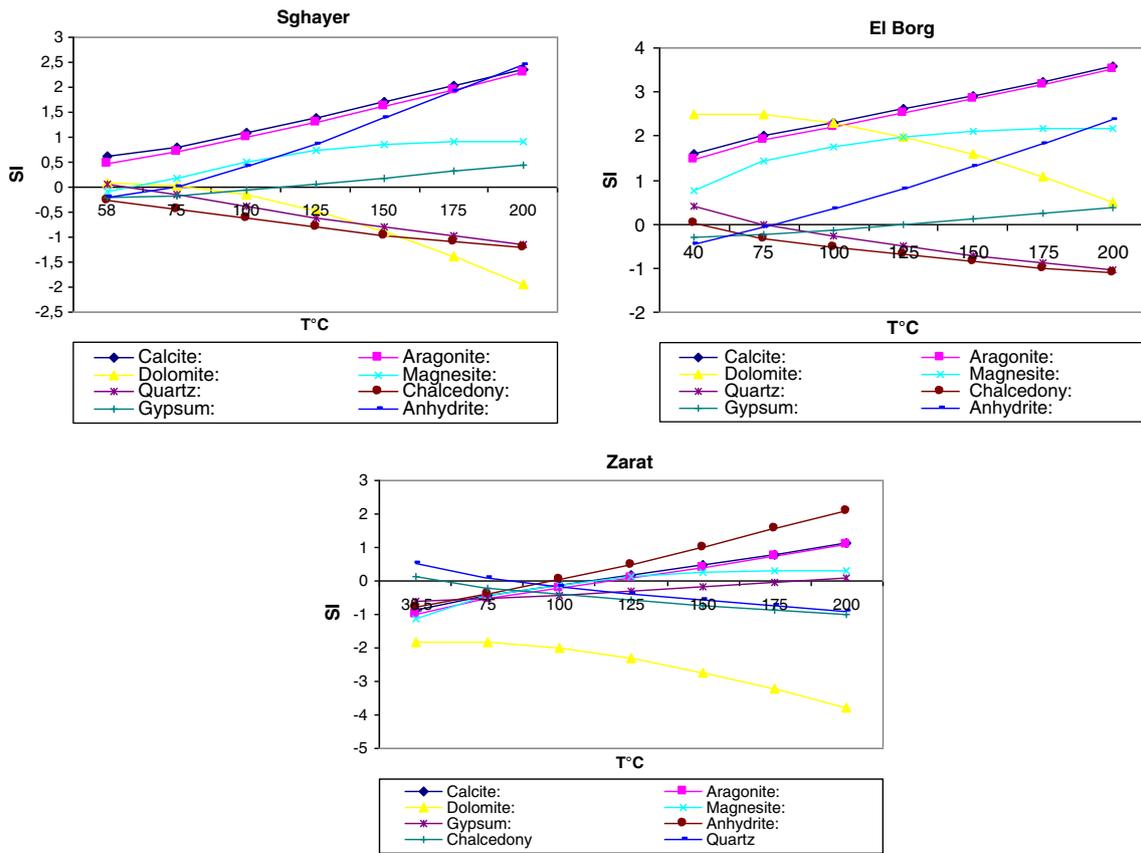


Fig. 12 Mineral saturation curves for the CI thermal water

Mixing of thermal and cold waters

Mixing processes are traced by chemical present in the mixed water and two end members (cold and thermal water). The temperature of the thermal water and the proportion of the two types of water in mixing can be estimated based on the chemical contents of cold and mixed water samples, using the method of Gupta and Roy (2007) involving two unknown equations. The first equation relates the enthalpies of the thermal water

(H_h), cold water (H_c) and spring water (mixing water) (H_s) and the fractions of cold water, X , and of thermal water ($1-X$).

$$H_c X + H_h(1 - X) = H_s \tag{1}$$

The second equation relates the silica contents of thermal water (Si_h), cold water (Si_c), and spring water (Si_s):

$$Si_c X + Si_h(1 - X) = Si_s \tag{2}$$

Table 6 Enthalpies of liquid water, and quartz solubilities at selected temperatures and pressures (Fournier and Truesdell 1974)

Temperature (°C)	Silica (mg/L)	Enthalpy (cal/g)
50	13.5	50
75	26.6	75
100	48	100.1
125	80	125.4
150	125	151
175	185	177
200	265	203
225	365	230.9
250	486	259.2

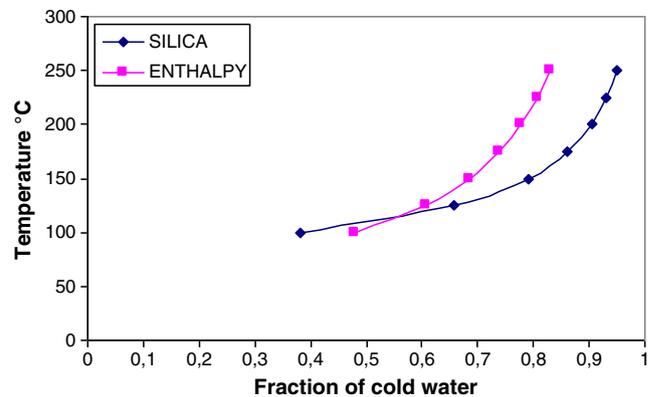


Fig. 13 Relation between fraction of cold water and temperature for Zarat spring

These two equations can be solved for the two unknowns. Here, a graphical approach suggested by Fournier and Truesdell (1974) is used for determining the unknowns in the spring water of Zarat as representative of the studied thermal waters (Gupta and Roy 2007):

The list of enthalpy as a function of temperature for pure water (Fournier and Truesdell 1974) is indicated in Table 6. The fraction X_t is calculated for each temperature value as follows:

$$X_t = H_h - H_s/H_h - H_c \quad (3)$$

Then X_t is plotted vs. the temperatures (T).

A series of values of the silica contents of thermal water for the temperatures (Table 6) (Fournier and Truesdell 1974) is considered and X_{Si} is calculated for each value as follows:

$$X_{Si} = Si_h - Si_s/Si_h - Si_c \quad (4)$$

X_{Si} is plotted vs. temperature on the same graph. The point of intersection of these two curves gives the estimated temperature of the thermal water component and the fraction of the cold water.

The mixing of different type of waters, resulting from the exchange between different aquifers, especially between the CI and the SI aquifers, was corroborated. In Zarat spring, considered as mixed water, the intersection of their two curves (Fig. 13) shows that the temperature of the thermal water is inferred to be less than 110 °C and the average contribution of cold water is about 57 %.

Conclusions

The south east Tunisian waters indicate different chemical composition related to different hydrogeological systems. The water of the two identified hydrogeological systems is characterised by different temperatures. The CI water is classified as thermal water (temperature of about 68 °C), whereas the water of the Djeffara Complex is classified as cold water (temperature of about 23 °C). In order to determine the geochemistry of these waters, the application of the Piper diagram reveals two types of facies: Na–Cl for the CI water and Ca–SO₄ for the cold water. With the projection of samples in the PCA approach, we have identified the line of mixing between the two types of water.

Using the silica geothermometers, the estimated temperature of the thermal reservoirs could be in the range of 50–87 °C, which is relatively close to the temperature estimated using equilibrium modelling (75–100 °C). The water temperature varies considerably suggesting mixing of ascending thermal water with shallow cold water. The contribution of cold water in mixing is estimated to be about 57 %.

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