

Geochemical study of groundwater mineralization in the Grombalia shallow aquifer, north-eastern Tunisia: implication of irrigation and industrial waste water accounting

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Received: 25 March 2008 / Accepted: 20 August 2008 / Published online: 9 September 2008
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Abstract Hydrochemistry of groundwater is largely determined by both natural processes, such as dissolution, cation exchange, mixing, evaporation; and anthropogenic activities, which can affect the aquifer systems by contaminating them or by modifying their hydrological cycle. Both natural and anthropogenic processes vary in time and space; which is reflected in groundwater hydrochemistry variation. The objective of this study is the determination of the main hydrogeochemical processes that affect the quality of shallow groundwaters in the Grombalia basin, located in the Cap Bon Peninsula, north-eastern Tunisia. In this area, the chemical composition of groundwater is mostly characterized by Na–Cl–NO₃–Ca water type which reveals the implication of natural and anthropogenic major factors. Natural factors are dissolution of evaporitic minerals, i.e. halite and gypsum and cation exchange with clays, while anthropogenic factors are pollution with industrial Sr-rich waste water and return flow of irrigation water, highly contaminated by MgSO₄ and methyl-bromide fertilizers.

Keywords Na–Cl–NO₃–Ca water type · Dissolution · Cation exchange · Industrial waste water · Irrigation return flow · Fertilizers

Introduction

The Grombalia basin, which extends over about 719 km², is located in the Cap Bon Peninsula, north-eastern Tunisia, between the latitudes 40° 59' and 40° 78' N and the longitudes 9° 02' and 9° 22' E. It is limited in the east by the Tekelsa hills, Abderrahman Mountain and the oriental coastal plain, in the south by the Hammamet hills, in the south-east by the Bouchoucha Mountain, in the east by the Halloufa Mountain and in the north by the Gulf of Tunis (Fig. 1). The climate of the region is semi-arid with a mean annual precipitation of 500 mm, mean annual temperature of 18.7°C and potential evapotranspiration of 1,187 mm/year. The surface water network in the study area is relatively dense due to the high amount of annual rainfall and the relatively low annual evapotranspiration. This network is made by El Bey non-perennial wadi and its tributaries, namely the Sidi Said, Tahouna, El Menzeh, Sidi Toumi, Bou Argoub and Masri wadis. These wadis collect surface runoff mainly from the southern and western foothill zones of the Bou Choucha and Bou Argoub mountains.

Urban, agricultural and industrial expansions in the province (Fig. 2), especially in the Soliman, Grombalia and Menzel Bou Zelfa regions, have caused an ever growing need for fresh water and have largely influenced the shallow groundwater reservoir quantitatively and qualitatively. The expansions, which began since the late 1970s and intensively increased over the 1980s and the 1990s due to the high regional potentialities for agriculture and industry, provoked a steady increase in groundwater withdrawal and water quality degradation. This has raised concerns relating to the effects on groundwater resources since (1) the rate of replenishment of the shallow aquifer is not known with precision and (2) the acute environmental problems related

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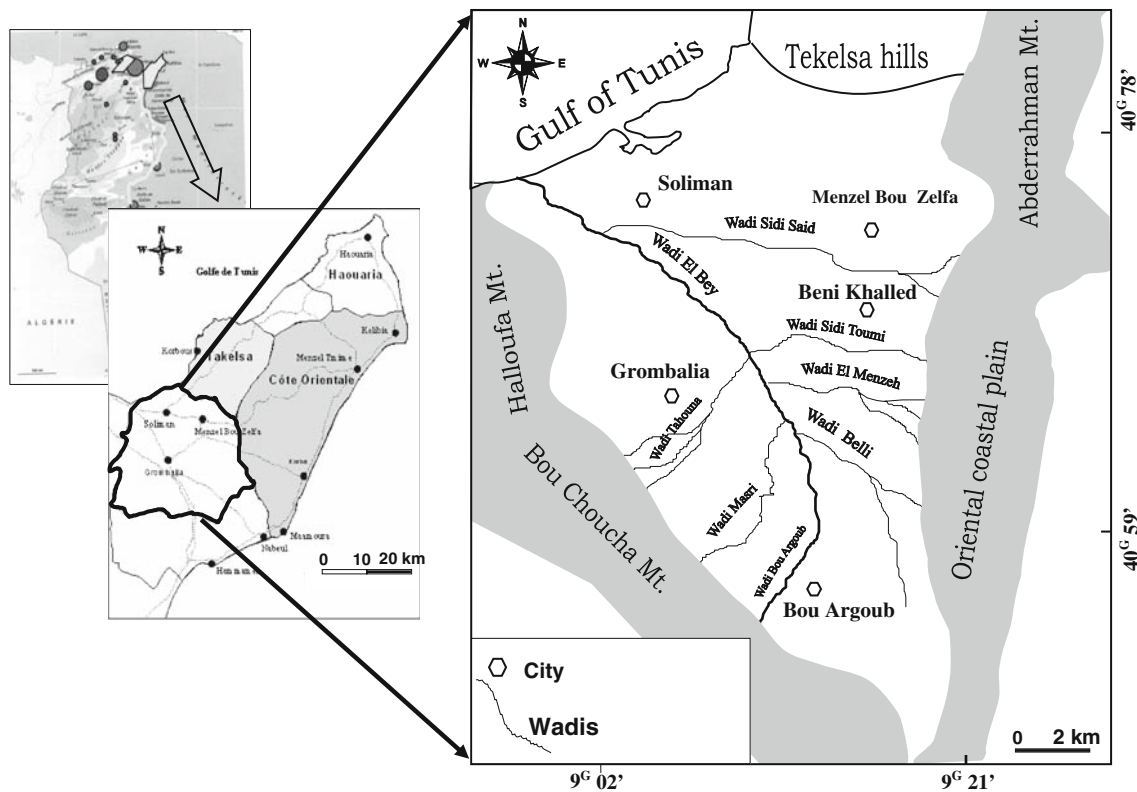


Fig. 1 Location map of the study area

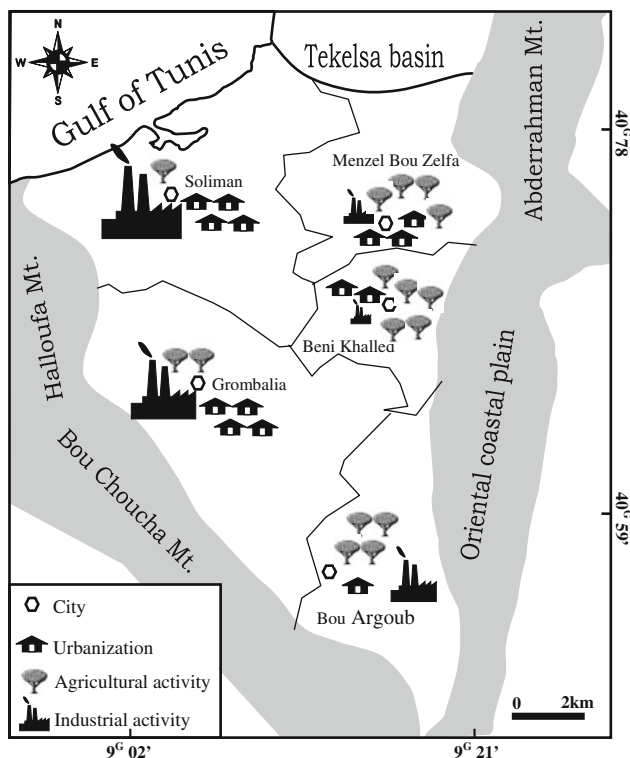


Fig. 2 Different districts and dominant land use activities in the Grombalia basin

to the groundwater pollution and salinization, which put a strain on the existing fresh water that supports the regional development.

Within this framework, this paper presents an attempt to identify the origins of groundwater salinization in the shallow aquifer. It also investigates the impact of regional agricultural and industrial activities on groundwater quality. The results will lead to a better management of groundwater resources in the Grombalia basin for viable long-term development in the region.

Geology

The study area is described as small sedimentary basin, whose edges appeared to be related to important flexures and normal faults, namely the Borj Cedria NNW–SSE fault and the Hammamet NE–SW fault, that have played after the Pliocene (Castany 1948). This basin is also known as a graben structure associated with normal faults having played in the Middle Miocene (Ben Ayed 1993; Chihi 1995).

As shown by the geological map (Fig. 3), the oldest formations in the area are Oligocene sandstones that belong to the Fortuna formation and Middle Miocene Sandstones and clays that constitute the Saouf formation. These two formations outcrop in the west of the Borj Cedria fault and

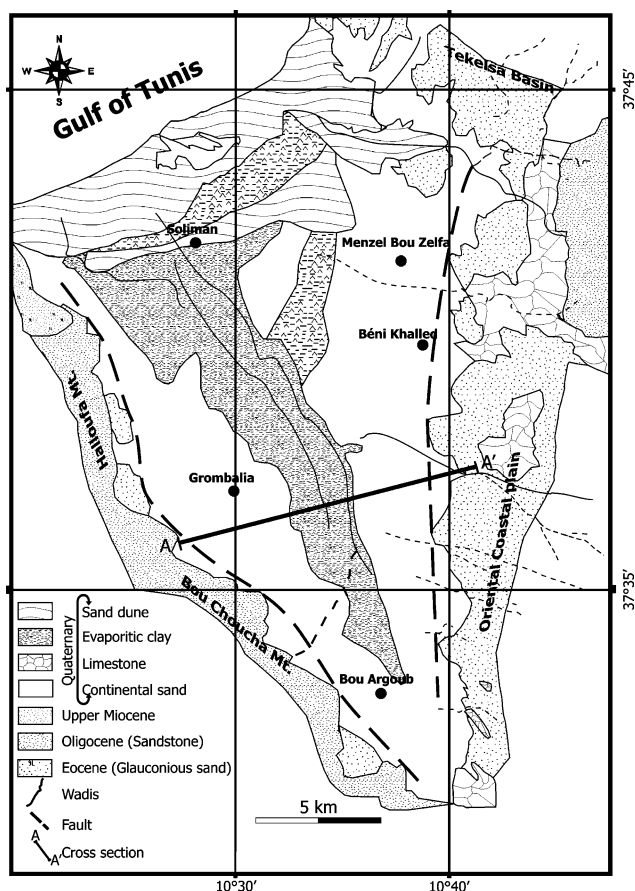


Fig. 3 Simplified geological map of the Grombalia basin

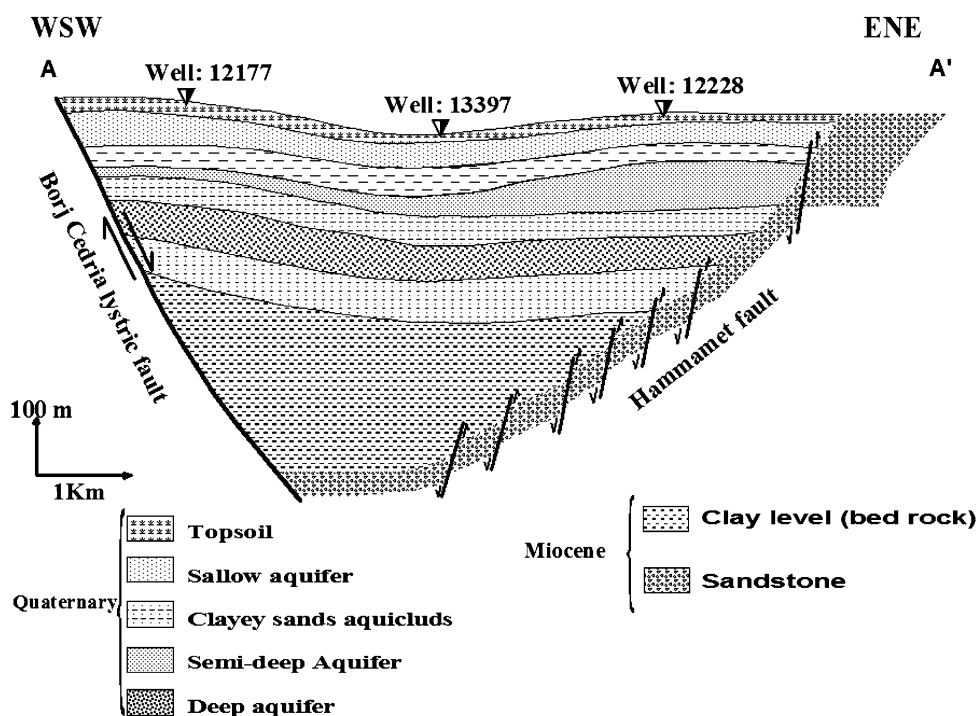
in the east of the Hammamet fault respectively (Ben Salem 1995). In the depth, the Grombalia graben is filled by fluvial Quaternary sediments with a thickness up to 430 m. These recent Quaternary deposits overlay uncomfortably the Miocene subsiding bed rock of the Saouf formation which itself normally reposes on the Fortuna sandstone formation.

The sedimentary units of Grombalia graben-filling, overlaying the Miocene bed rock, consist mainly of detrital sediments of sandy clays and silts, fine to coarse grained sands and some sandstone deposits. This sedimentation is intercalated with some thick clay levels and lenses. Some evaporites consisting mainly of halite and gypsum and/or anhydrite minerals characterize especially the central part of the basin.

Hydrogeology

As shown in the hydrostratigraphic cross section (Fig. 4) the Grombalia shallow aquifer is represented by an intercalation of several Quaternary levels which constitutes the filling of the Grombalia graben. Hydrogeologically, the tow faults of Borj Cedria and Hammamet that contributes to the existence of the shallow aquifer formations have largely influenced the hydrodynamic functioning of the water table. Indeed, these faults have played the role of subsurface barrier contributing to the hydraulic discontinuity of the Grombalia aquifer in one hand; and the aquifer systems of the Zaghouna fluvial plain

Fig. 4 Hydrogeological cross section in the Grombalia basin



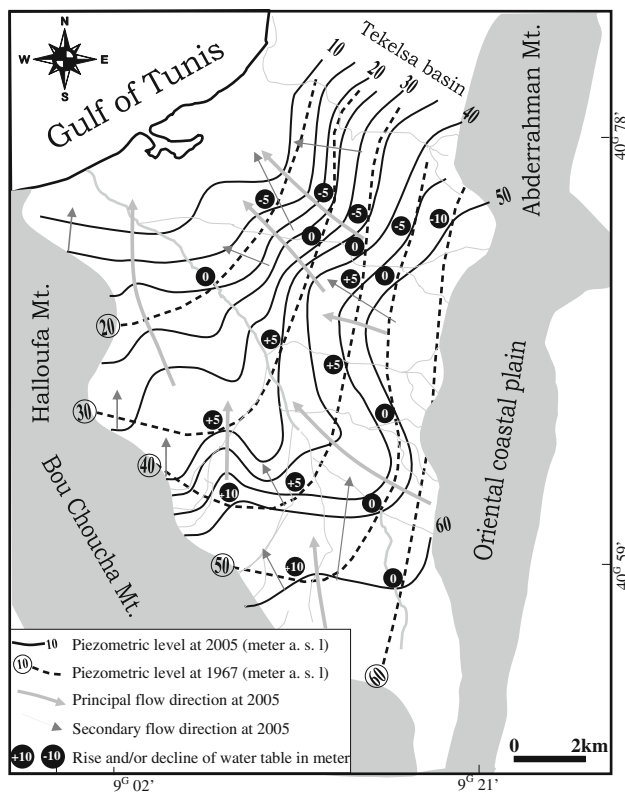


Fig. 5 Piezometric map of the shallow aquifer water table at 2005 and 1967 showing the evolution of the groundwater level

(in the south-west) and the oriental costal plain (in the south-east) in the other hand.

The hydraulic conductivity and the transmissivity of the Grombalia shallow aquifer present a large range of variation due to the changes in grain size and local increases of clay deposit.

The piezometric map of the shallow aquifer water table in the study area (Fig. 5) was established on the basis of water level measurements of 41 wells realized in March 2005. This map shows that the main groundwater flow direction in this basin is SE-NW, from Bou Argoub foot hill zone towards the Gulf of Tunis which constitutes the natural discharge area. Another main flow direction appears in the north-eastern part, from the Menzel Bou Zelfa hills towards the El Melah depression. The two flow directions highlight the importance of rainfall infiltration in Bou Argoub and Menzel Bou Zelfa areas. Secondary flow directions arise from the bordering hills highlighting their important contribution in the recharge of the Grombalia shallow aquifer. Nevertheless, in spite of the existence of several important streams, the linear recharge type is insignificant, except in the north-eastern part of the basin. However, the spatial recharge type plays an important role because of the important yearly rainfall amount and the highly permeable Quaternary deposits which outcrop in most parts of the basin.

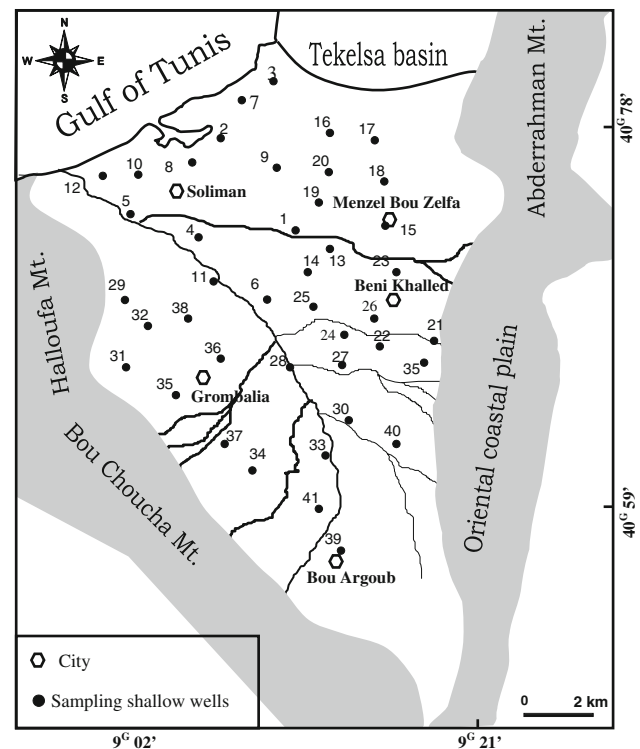


Fig. 6 Sampling map

On the other hand, the comparison between the piezometric map realized within the present study and that established 40 years ago (Castany 1967), before the development of the land use activities in the Grombalia basin, confirm the importance of the spatial recharge in the most parts of the basin. It also indicates that the irrigation by the treated waste water plays a significant role in the alimentation of the shallow water table, especially in the zones where the industrial activity is more developed. Indeed, in spite of the important withdrawal, the water table has registered a remarkable rise varying between 0 and 10 m in the whole basin, except for the north-eastern part of the Menzel Bou Zelfa area in which the groundwater level has show an important decline. This decline can be explained by the intensive over-pumping in relation with the important expansion of agricultural activity in this area.

Sampling and analytical procedure

A field sampling campaign was carried out during September 2005. A total of 38 groundwater samples was collected from dug wells tapping the shallow aquifer at depths ranging between 6 and 35 m (Fig. 6). In situ measurements of pH, Temperature, electric conductivity (EC)

Table 1 In situ measurements and geochemical data of Grombalia shallow groundwaters

Sample number	T (°C)	pH	CE (mS/cm)	TDS (mg/l)	Cl (meq/l)	Na	Ca	SO ₄	HCO ₃	Mg	Sr	Br	NO ₃	SI halite
1	17	7	5.72	4,738	45.2	39.0	19.6	17.5	7.0	13.4	1.1	2.0	2.4	−4.391
2	18.3	7.29	5.22	4,139	48.1	38.1	17.1	11.8	4.0	9.7	0.8	2.5	5.0	−4.508
3	21.2	7.33	3.55	2,695	19.2	21.0	12.0	10.7	4.7	5.6	0.3		2.4	−5.141
4	12.9	7.86	5.01	3,834	33.4	30.9	11.8	14.7	4.1	13.5	1.1	1.4	0.0	−4.732
5	14.7	7.37	2.76	2,004	16.5	10.4	8.3	4.2	5.5	4.7	0.4	12.3	1.4	−5.47
6	16	7.34	4.89	4,221	42.4	30.3	30.2	14.7	4.9	11.9	0.9	1.7	6.2	−4.663
7	18.4	7.64	1.122	746	5.0	4.5	5.4	1.4	4.8	1.5	0.2	6.3	0.4	−6.319
8	17.6	7.47	2.81	1,958	12.1	13.4	9.8	7.5	6.2	4.8	0.5	2.5	1.2	−5.506
9	19.4	7.17	4.1	3,040	29.0	20.7	16.7	9.0	4.9	7.7	0.5	10.2	3.4	−4.974
10	19.5	7.28	3.7	2,673	22.6	20.3	9.9	7.4	4.8	5.6	0.7	2.2	0.5	−5.072
11	18.3	7.02	4.07	3,442	21.7	26.8	13.7	14.7	10.0	10.7	0.7	11.4	1.5	−4.989
12	17	7.53	1.821	1,292	9.5	9.3	5.9	4.1	3.8	4.2	0.3	4.2	1.0	−5.75
13	20.6	7.05	2.55	2,011	9.2	12.2	10.0	9.7	8.3	4.6	0.4	5.5	2.2	−5.677
14	18.1	7.28	3.91	3,230	16.9	15.8	17.3	19.8	6.8	12.1	0.9	1.1	0.6	−5.325
15	11.9	7.63	1.817	1,399	10.2	6.5	10.7	4.4	3.8	1.5	0.1		1.4	−5.861
16	18.3	7.16	4.35	3,618	17.9	17.5	22.8	21.0	6.4	5.1	0.4	9.1	0.8	−5.256
17	17.5	7.41	2.25	1,540	8.7	10.7	8.6	5.1	6.0	2.2	0.1	3.3	2.4	−5.733
18			2.22	1,705	12.5	6.3	10.8	2.3	4.3	2.5	0.1	5.9	1.9	−5.804
19	15.8	7.02	3.93	3,035	19.9	18.7	18.4	12.7	8.1	8.3	0.6	9.0	1.3	−5.172
20	16.8	7.43	3.27	2,474	18.6	14.8	18.5	8.7	6.3	4.1	0.2	7.2	1.3	−5.291
21	14.6	7.41	1.695	1,155	9.0	7.0	6.5	1.5	5.9	2.2	0.1	1.5	1.0	−5.877
22	16.9	7.07	4.65	3,578	29.9	23.2	19.3	9.7	6.4	8.6	1.4	1.4	1.6	−4.908
23			4.19	3,363	24.5	15.8	24.7	8.6	5.0	5.3		3.7	3.9	−5.16
24	17	7.09	7.84	5,914	66.9	44.2	27.5	11.6	4.7	20.8	1.2		5.8	−4.32
25	14.8	7.61	7.09	5,694	55.1	43.3	17.0	15.9	7.4	18.0	0.9	15.0	0.0	−4.396
26	14.8	7.33	3.54	2,949	17.9	10.0	21.2	10.8	5.9	6.6	0.7	13.1	2.5	−5.478
27	15.5	7.09	6.65	5,416	50.6	35.2	24.0	11.8	5.9	16.9	1.3	16.0	2.6	−4.522
28	16.5	7.37	4.38	3,493	20.7	23.9	12.1	17.7	8.0	13.4	1.6	11.9	1.2	−5.055
29	16.3	7.14	2.4	1,865	18.2	11.8	7.5	1.4	6.2	4.0	0.6	6.7	0.0	−5.374
30	13.9	7.23	5.6	4,730	50.3	37.5	19.0	17.5	6.9	16.7	1.0	2.9	1.1	−4.494
31	17.2	6.91	2.68	2,199	19.8	13.2	12.8	5.0	7.6	4.5	0.5	7.8	1.1	−5.305
32	15	7.19	2.77	2,082	19.2	14.8	10.3	3.3	6.4	4.5	0.6	6.2	1.5	−5.257
33	15.9	7.5	3.65	2,995	15.0	19.2	10.1	13.2	8.8	9.1	0.0	6.0	2.2	−5.279
34	16.4	7.04	3.7	2,953	18.7	17.1	13.4	11.3	6.2	7.4	0.5	9.0	1.0	−5.229
35	16.6	7.17	3.08	2,258	15.2	11.4	16.1	9.1	7.5	7.5	0.3	5.2	0.8	−5.488
36	16.2	7.53	2.57	1,998	22.8	14.7	9.1	3.3	6.0	6.0	0.4	11.2	0.0	−5.19
37	16.4	7.25	3.72	3,034	24.8	20.4	16.9	8.7	6.8	7.4	0.3	9.7	1.8	−5.804
38	13.4	7.61	5.18	3,926	29.8	34.6	10.5	12.1	5.7	12.3	0.8	19.4	4.9	−5.677
39	14.5	7.21	2.77	2,057	14.9	13.4	13.9	8.7	6.5	4.1	0.2	7.1	2.1	−5.419
40	17.6	7.92	1.419	970	7.2	6.0	5.1	3.2	4.6	3.2	0.2	2.9	0.2	−6.047
41	16.8	7.02	4.16	3,517	24.3	15.8	19.9	14.2	6.4	11.4	0.8	1.3	0.9	−5.164

and the total dissolved solids (TDS) were performed in the field. Water samples were filtered and collected in 100 ml polyethylene bottles with poly-seal caps for major and minor elements analysis which have been done at the

“Laboratoire de Radio-Analyses et Environnement” of the “Ecole Nationale d’Ingenieurs de Sfax” (Tunisia), using titration method (for HCO₃) and standard ion chromatography techniques.

Results and discussion

In situ measurements interpretation

In situ measurements of temperature, pH, TDS and EC in groundwater samples are shown in Table 1. Groundwater samples of different regions of the Grombalia basin show relatively heterogeneous values of temperature, varying between 13.9 and 21.2°C. Relatively high temperatures, close to that of the atmosphere, characterize Soliman and Menzel Bou Zelfa regions. These high temperatures are probably related to the shallow depth of the groundwater table. However, deeper wells of Grombalia, Beni Khaled and Bou Argoub regions are marked by relatively low temperatures, indicating that they are not or slightly influenced by the atmosphere. Inexpectly, groundwater samples are almost neutral. Their pH values are very homogeneous and present a narrow range of variation (between 6.9 and 7.8), which does not reveal any contamination neither by domestic and industrial waste water, produced in Soliman and Grombalia zones, nor by return flow from irrigation water characterizing Beni Khaled and Bou Argoub agricultural regions. Values of EC vary largely between 1.1 and 7.8 mS/cm probably due to the interference of several natural and anthropogenic

mineralization processes, i.e. dissolution and pollution with fertilizers or industrial waste waters.

Groundwater salinity, represented by the TDS values, shows a large range of variation from <1 g/l to almost 6 g/l. This variation conforms partially with the main groundwater flow directions, indicating that the groundwater salinity is somehow controlled by the residence time in the aquifer. Indeed, relatively low TDS values characterize the eastern, southern and western bands of the study area reveal the dilution of the groundwater by the recharge coming from the basin border highlands. However, high salinities appear in the central part of the basin which relatively disturbs the evolution of the mineralization in the direction of the groundwater flow. This sudden increase of groundwater salinity is expected since the central part of the study area is marked by the abundance of evaporitic deposits and the development of land use activities.

Major elements geochemistry

Water types

Major ions of groundwater are plotted on the Piper trilinear diagram (Piper 1944) in order to distinguish the different

Fig. 7 Piper diagram of the shallow aquifer groundwaters

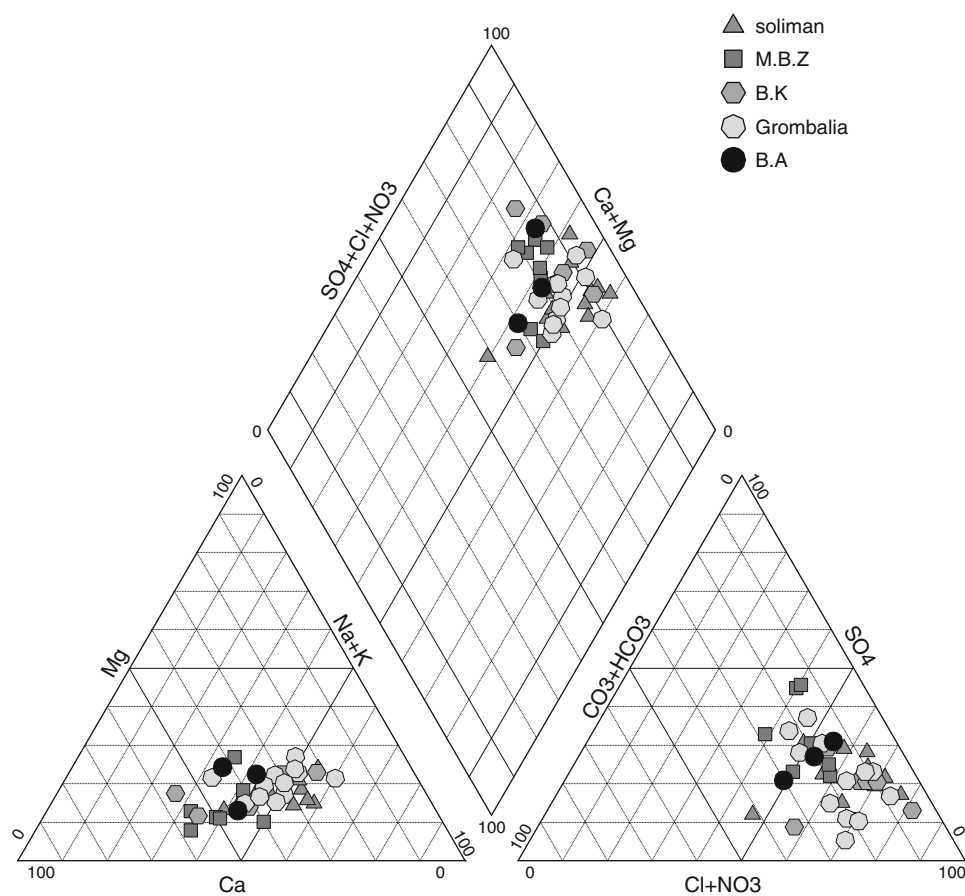
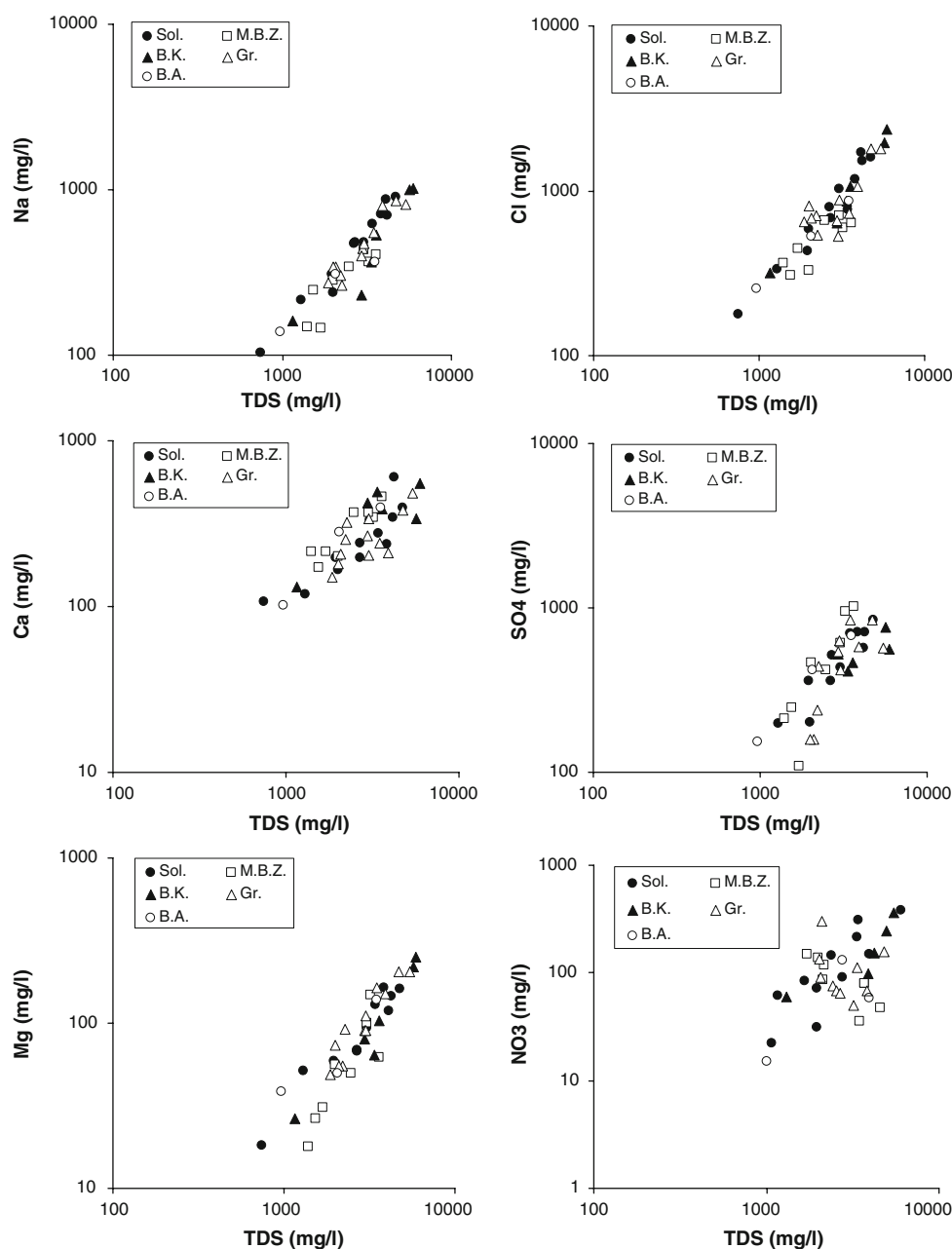


Fig. 8 Major elements and nitrate versus TDS relationships



water types in the Grombalia shallow aquifer. Nitrate component was taken into account when plotting this diagram because of its relative abundance in the groundwater (Fig. 7). Samples are classified according to the geographical district from where they are collected in order to identify whether the water type is influenced by land uses of each district. In the Piper diagram all samples show the same Na–Cl–NO₃–Ca water type. This probably indicates the ineffectiveness of major elements concentrations in identifying the influence of land use activities on groundwater mineralization since such effect is mostly masked by the dissolution process.

Groundwater mineralization processes

Bivariate diagrams of major elements versus TDS values are used to identify geochemical processes that contribute to the groundwater salinization in the Grombalia shallow aquifer (Fig. 8). As shown in these diagrams, well-defined correlations characterize the relationship of Na, Cl, Ca, SO₄ and Mg versus TDS values. These positive correlations indicate that the referred elements contribute to the groundwater mineralization. Indeed, spatial distributions of the mentioned elements and TDS values show practically the same evolution (Fig. 9).

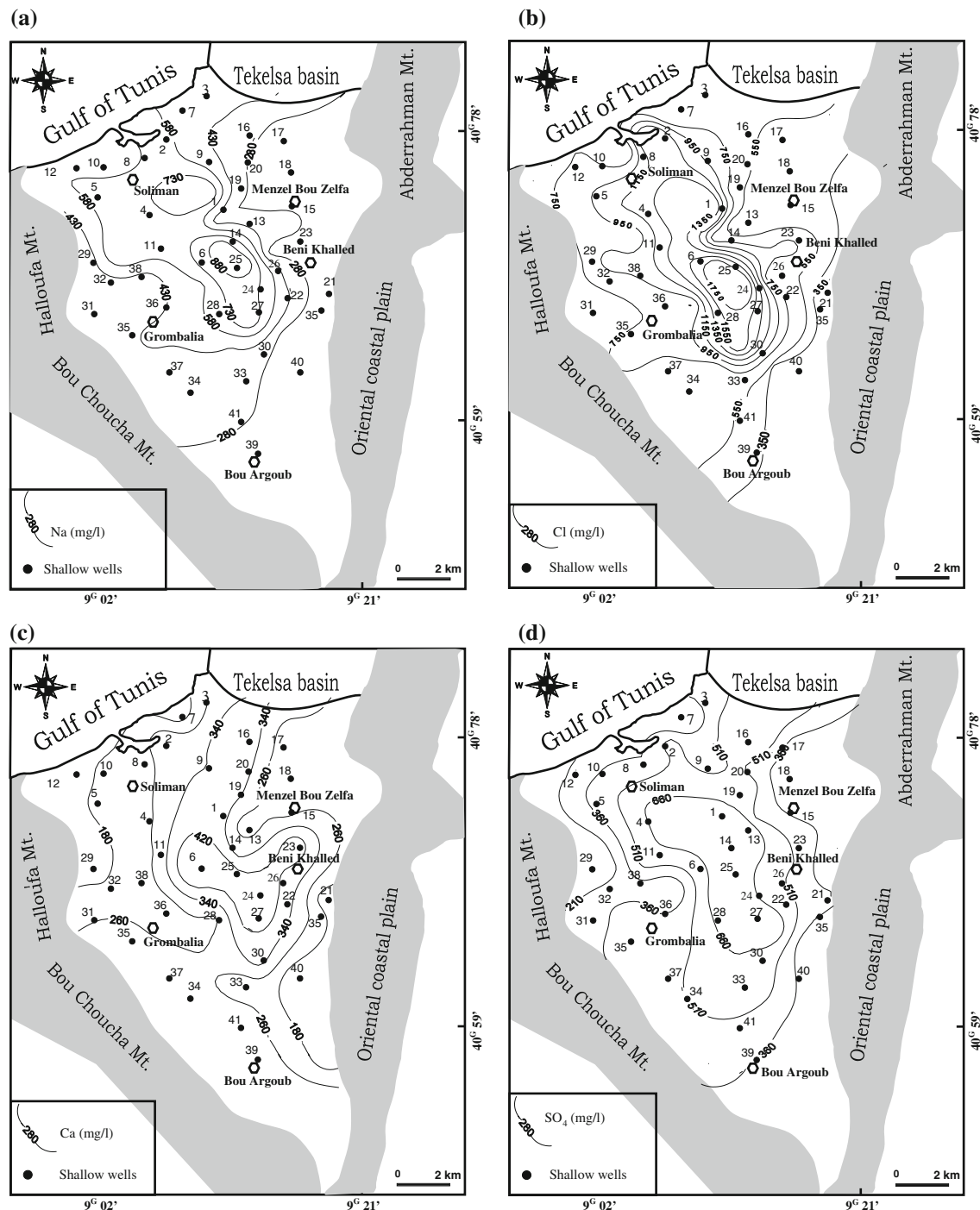


Fig. 9 Spatial distributions of Na (a), Cl (b), Ca (c) and SO_4 (d)

In order to specify the likely origin of the each major element participating to the groundwater salinization, plots of Na versus Cl, Ca versus SO_4 and Mg versus SO_4 are established. The first plot displays a well-defined correlation (Fig. 10) indicating that Na and Cl ions have probably the same origin, which is the likely dissolution of halite deposits relatively abundant particularly in the central part of the basin. This dissolution is, however, confirmed by the

negative saturation indexes of groundwater with respect to halite which indicates an undersaturation state. Moreover, these saturation indexes vary in inverse proportion to the some of Na and Cl concentrations (Fig. 11). The second plot of Ca versus SO_4 shows a moderate correlation (Fig. 12), indicating that the dissolution of gypsum and/or anhydrite is relatively limited. It characterizes mainly the Soliman region located in north-western part of the basin.

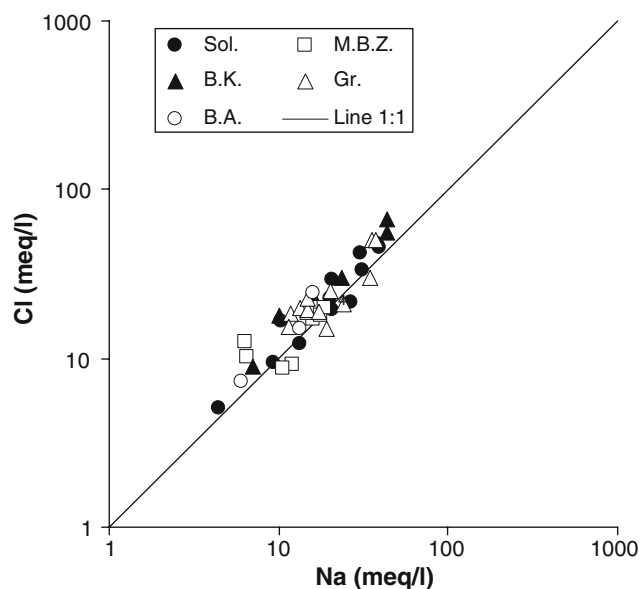


Fig. 10 Na/Cl relationship

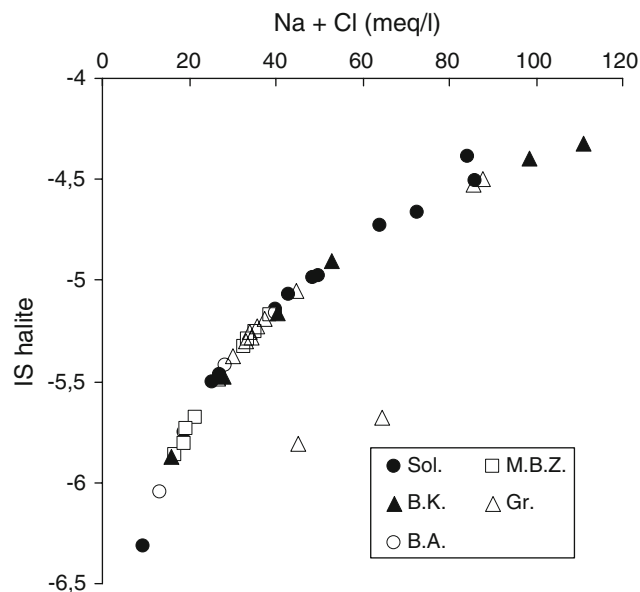


Fig. 11 (Na + Cl)/IS (halite) relationship

The third plot of Mg versus SO_4 (Fig. 13) exhibits a well-defined relationship which indicates an origin of Mg and SO_4 related to either the dissolution of MgSO_4 evaporitic minerals (Adams et al. 2001) or the contribution of MgSO_4 -fertilizers (Gi-Tak et al. 2004), commonly used in the Beni Khaled and Menzel Bou Zelfa agricultural areas.

On the other hand, in the plot Na/Cl a relative deficiency of Na^+ with respect to Cl^- is observed. This deficient is probably due to the cation exchange process that typically characterizes clay minerals-rich aquifers. Indeed, this deficiency of Na in the relationship Na/Cl is compensated by an excess of Ca^{2+} with respect to SO_4^{2-} . Moreover, the

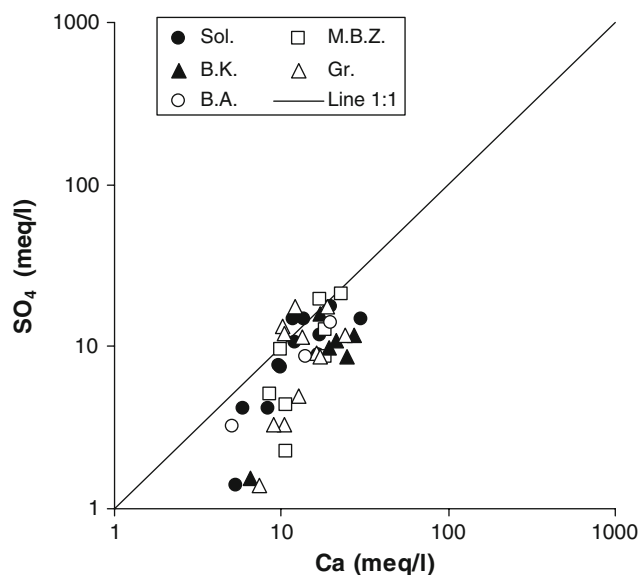


Fig. 12 Ca/ SO_4 relationship

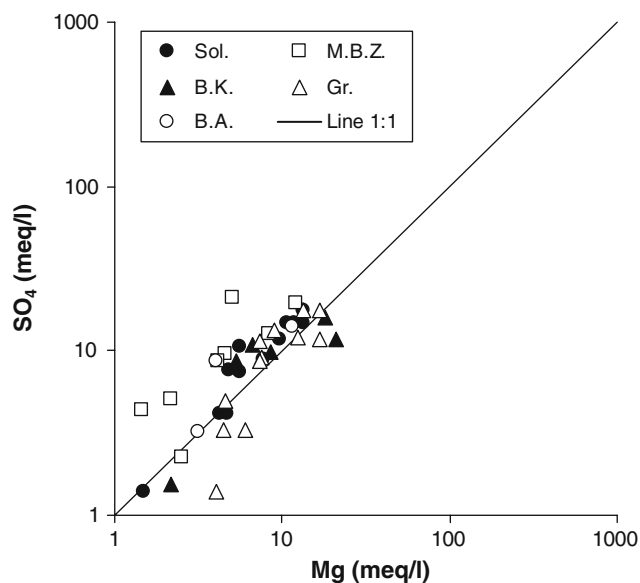


Fig. 13 Mg/ SO_4 relationship

referred cation exchange is confirmed through the relationship illustrated in Fig. 14 in which $(\text{Na} - \text{Cl})$ and $[(\text{Ca} + \text{Mg}) - (\text{SO}_4 + \text{HCO}_3)]$ vary in inverse proportion (Garcia et al. 2001). In the absence of this exchange all data should plot close to the origin (Mc Lean et al. 2000).

Nitrate and trace elements geochemistry

Nitrate

Nitrate values in the Grombalia shallow aquifer are relatively high. They vary in a wide range, between 0 and

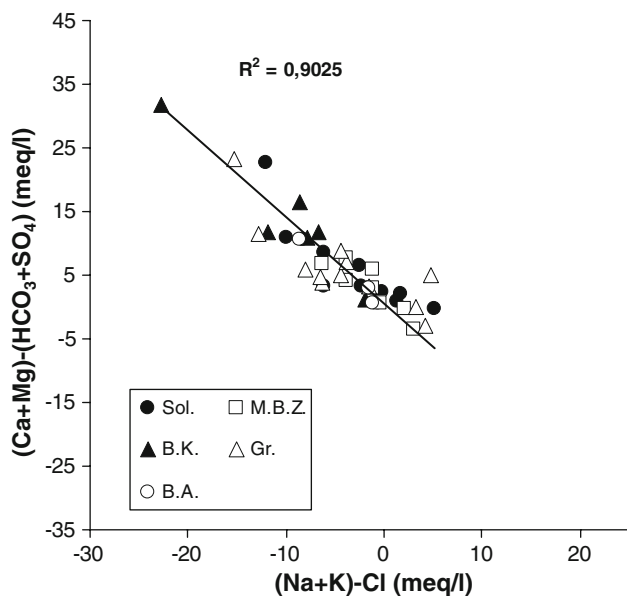


Fig. 14 $(Ca + Mg) - (SO_4 + HCO_3)/(Na + K - Cl)$ relationship

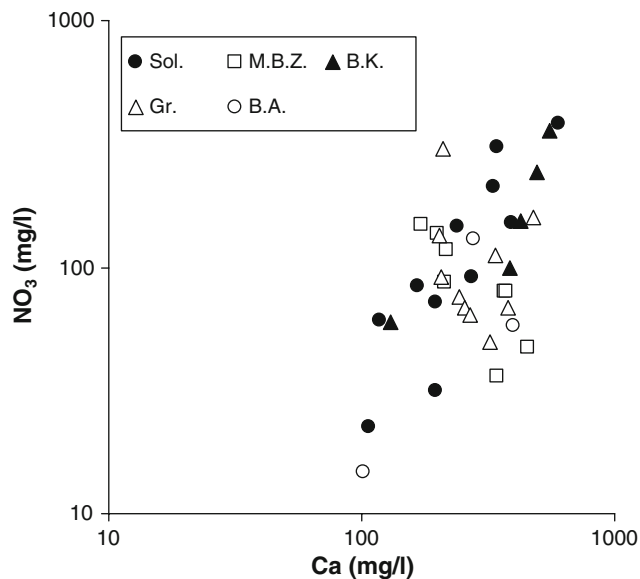


Fig. 16 Ca/NO_3 relationship

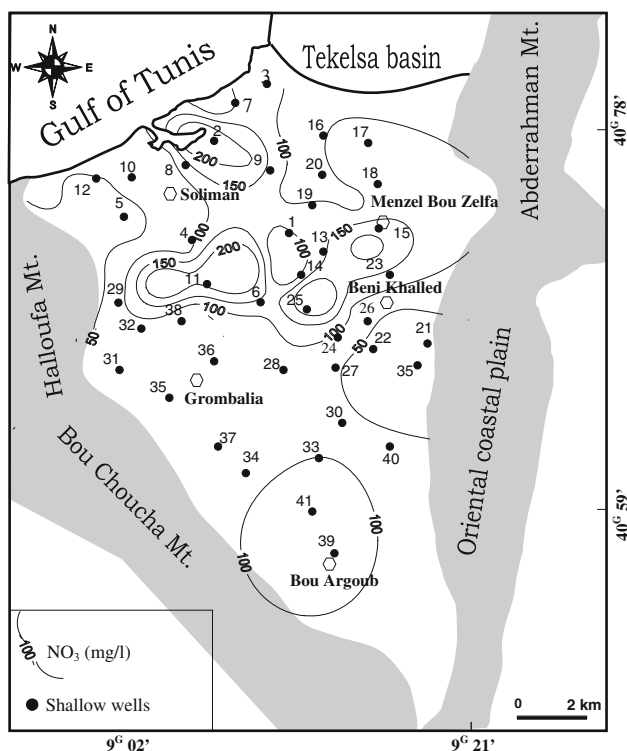


Fig. 15 Map of nitrate distribution

384 mg/l. In 32 wells, located mainly in the central part of the basin, nitrate concentrations exceed 50 mg/l, which represents the maximum european admissible nitrate concentration in drinking water. The spatial distribution of nitrate concentrations, represented in Fig. 15, clearly indicates that the high contents are related to the agricultural

land use patterns. This suggests that the application of nitrogen fertilizers leads to increased nitrate leaching. The areas with nitrate concentrations >50 mg/l are located in the Soliman and Beni Khaled region where agricultural activities are more developed. In deed, the positive relationship between NO_3 and Ca (Fig. 16) suggests that both elements are utilized in the highly polluted regions of Soliman and Beni Khaled as $Ca(NO_3)_2$ -fertilizers (Stigter et al. 2006).

On the other hand, high nitrate concentrations highlight the significant role of the recharge by return flow from irrigation water in the mentioned regions where flood irrigation is particularly used. Besides, in these regions the NO_3 contents in the groundwater correlate positively with the TDS values indicating the significant contribution of this element in the salinization process (Fig. 8).

Trace elements

Strontium and bromide trace elements are inexpectly abundant in the groundwater of the Grombalia shallow aquifer. Such elements are generally useful in identifying the mixing effect between groundwater bodies of different aquifer levels, especially when they are used in conjunction with other conservative elements such as chloride. In the Grombalia shallow aquifer, these elements are utilized to identify sources of groundwater pollution as well as hydrogeochemical reactions accompanying salinization processes.

Localized high strontium concentrations (>2.5 mg/l) occur in the Grombalia region, central-west of the basin. However, moderate Sr contents characterize Soliman and

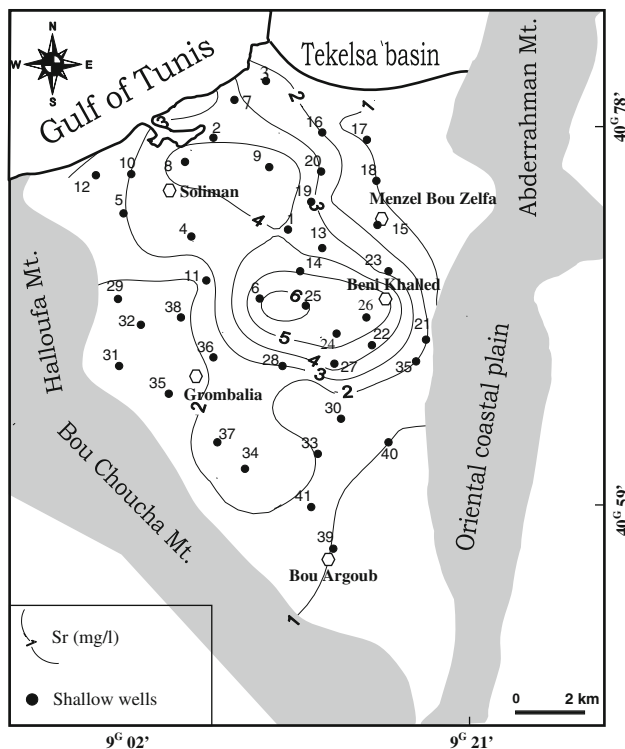


Fig. 17 Map of strontium distribution

Beni Khaled regions, situated in the centre of the basin, and relatively low contents distinguish Menzel Bou Zelfa and Bou Argoub regions, located respectively in north-east and the south-east of the study area (Fig. 17).

High and moderate strontium concentrations in the Grombalia, Soliman and Beni Khaled regions are probably due to both celestite (SrSO_4) dissolution, as shown by the positive correlation between Sr and SO_4 illustrated in Fig. 18, and downward leakage in relation with return flow from irrigation with treated industrial waste water. In fact, this return flow is strongly suggested considering the relatively low depth of the shallow aquifer water table in the mentioned regions. In the Menzel Bou Zelfa and Bou Argoub zones, the ratio of strontium to sulphate is in disagreement with the celestite dissolution (Fig. 18). Likely source of Sr in these regions is the discharge of industrial non treated waste waters particularly in the Bou Argoub area where these waters are directly discharged into the surface drainage network.

In general, the content of bromide in groundwaters is low, with an average value of 0.00056 mmol/l, i.e. 0.0447 mg/l (Hitchon et al. 1999). Nevertheless, high concentrations of this element sometimes occur in groundwater by pollution, interaction with seawater or reaction with evaporate deposits (Richer and Kreitler 1993). In the study area, waters located in the Grombalia region present the highest bromide concentrations with an average of 9.3 mg/l. Moderate

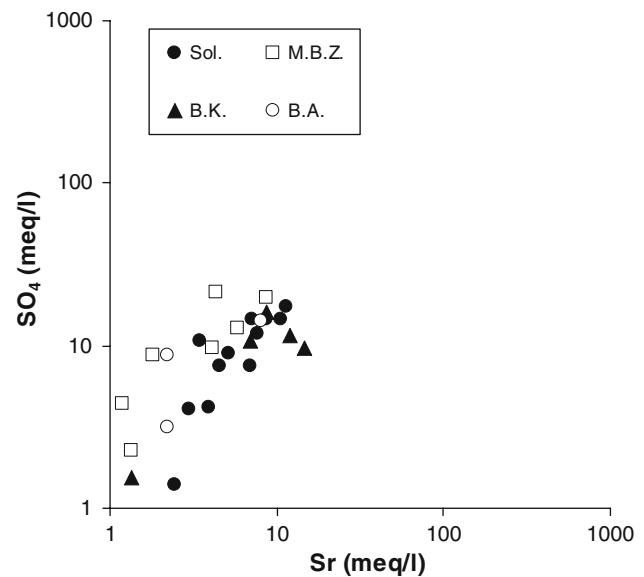


Fig. 18 Sr/ SO_4 relationship

concentrations with average values of 5.1, 5.8 and 6.9 mg/l are measured in the Soliman, Menzel Bou Zelfa and Beni Khaled regions, respectively. However, low concentrations characterize the Bou Argoub region with an average of 3.7 mg/l. Logically, such bromide concentrations are too high to be derived from evaporate sources because, when existing in salts, bromide is usually a trace element that does not exceed 1 mg/l. Moreover, marine origin of bromide is also excluded for the reason that high Br concentrations are localized in the central part of the basin (in Grombalia and Beni Khaled) but not in the Soliman coastal region. Thus, likely origin of these high Br concentrations in the study area is groundwater pollution by return flow from irrigation water, which contains high amounts of methyl bromides and other organic bromyl pesticides (Zhu et al. 2007).

Conclusion

The combination of physical hydrogeologic tools and the hydrogeochemical tracers, i.e. majors and minors elements permits to understand the hydrodynamic functioning of the Grombalia shallow aquifer. The potentiometric map indicates the role of spatial recharge type in the whole basin and in the bordering hills in the alimentation of the shallow water table.

Salinization process and contamination of shallow groundwaters in the Grombalia basin were investigated in relation with industrial and agricultural land use. This investigation shows a very variable chemical composition of groundwater, e.g. EC varies in wide range between 1.1 and 7.8 mS/cm. The hydrogeochemical characteristics of groundwaters, which comprise a main $\text{Na-Cl-NO}_3\text{-Ca}$

water type, are affected by four natural and anthropogenic salinization factors namely, the dissolution of halite and gypsum, the Na–Ca cation exchange, the pollution by industrial Sr-rich waste water and the return flow of irrigation water contaminated by MgSO_4 and methyl-bromide fertilizers.

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