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Impact of water resources management on groundwater hydrochemical changes: case of Grombalia shallow aquifer, NE of Tunisia

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Abstract

Grombalia region (NE Tunisia) exploits multiple sources of water: groundwater from shallow and deep aquifers, local surface water, and external surface water transferred from the North-west of Tunisia (Medjerda River and Ichkeul basin). The coordinated control of the water sources is very week while no evaluation of the impact of anthropogenic activities on groundwater resources was carried out so far. The present study use water-level time series, rainfall, and groundwater extraction data to identify the piezometric changes. Geochemical data were used to characterize and classify water samples and to study water–rock interaction based on ion plots and diagrams, mineralization, and nitrate contamination processes, as well as their fate and origins. The lack of regulation of the water use is potentially responsible of the increase of both level and salinity of groundwater in the central and downstream parts of the basin and of a large groundwater drawdown in the upstream region.

Keywords Groundwater rise · Salinization · Nitrate contamination · Anthropogenic activities · Geochemical multi-tracer · Return-flow

Introduction

In arid and semi-arid regions, groundwater represents a crucial water resource (Lachaal et al. 2012; Jellalia et al. 2015). The groundwater overexploitation is usually characterized by the decrease in groundwater levels (Lachaal et al. 2014; Massuel and Riaux 2017) and the degradation of the water quality (Kirby et al. 2015). Seawater intrusion and salinization of coastal aquifers are the major environmental problem caused mainly by overexploitation of natural groundwater resources (Chekirbane et al. 2014; Najib et al. 2016). In other cases, the intensive use of water for irrigation need without good drainage system and anthropogenic activities related to the urban and industrial wastewater discharge into rivers, can cause an increase of groundwater levels (Kirby et al. 2015; Yousif et al. 2016), salinazation (Mhamdi et al. 2015; Najib et al. 2016), and nitrate contamination (Walraevens et al. 2015; Askri 2015).

Previous chemical studies show the degradation of Grombalia groundwater quality (Ben Moussa et al. 2009; Lachaal et al. 2016, 2018). A high groundwater salinization is caused by dissolution of evaporitic rocks (halite and gypsum) (Charfi et al. 2013) and by the industrial and agricultural activities (Ben Moussa et al. 2009). Other previous works are interested to evaluate the water resources management strategies and its implications on hydrodynamic and hydrochemical changes in the Grombalia region (Lachaal et al. 2016, 2018). These studies show that the region is characterized by a poor water resources management. As consequence, groundwater rise was registered in the central part of the aquifer, causing many agricultural, environmental, and economic issues. Groundwater rise exceeds 2-m depth from the surface. In the same area an increase in water salinity and nitrate concentration were recorded (Lachaal et al. 2016; Lachaal et al. 2018).

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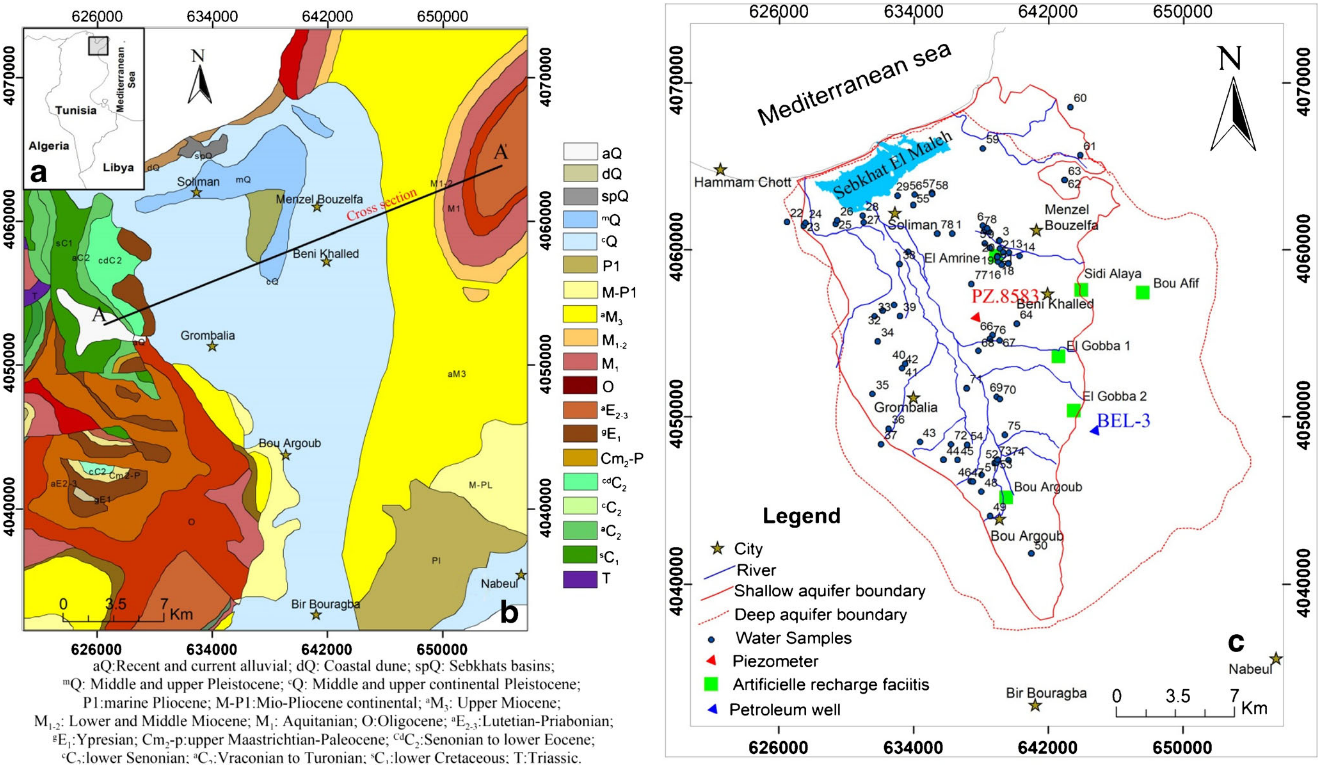
The aim of this study is to characterize the groundwater chemical properties and changes in Grombalia region, in relation to the water resources use and anthropogenic activities, and to identify the major hydrogeochemical processes occurring in Grombalia aquifer.

Geologic and hydrogeologic contexts

Studied area is located in Grombalia plain, in the North-east Tunisia, with a total area of 100 km^2, between 625,000 and 655,000 m North parallels and the 4,037,000 and 4,070,000 m East meridians (UTM32N, System) (Fig. 1). It is bounded to the North-east by Jebel Korbous, to the east by Jebel Abdelrrahman and Takelsa syncline, to the South-east by Hammamet–Nabeul plain and Jebel Reba El Aine, to the South-west by Jebel Halloufa and Jebel Bouchoucha, and to the North-west by the Mediterranean Sea (Fig. 1a, b).

Grombalia basin is formed by the Grombalia graben oriented NW-SE (Figs. 1b and 2). The graben is formed by two major normal faults: Borj Cedria fault from the South-West and Hammamet fault from the North-east. Grombalia basin is characterized by a strong subsidence of Beglia Formation (Miocene) and Segui Formation (Mio-Plio-Quaternary) (Chihi 1995; Hadj Sassi et al. 2006) (Fig. 1b). In the central part of the basin, the depth of Beglia and Segui Formations is variable due to the graben effect. It reaches a depth of 1000 m (Hadj Sassi et al. 2006). The schematic and conceptual model of Grombalia aquifer system A-A′ shows that aquifer system is limited to the East-north-east by Jebel El Hofra and to the West-south-west by Jebel Ghourfa, Jebel Djemaa, Jebel Messaoud, and Mornag aquifer system (Fig. 3) (Lachaal et al. 2016). The major geological outcrops in the study area are mainly Quaternary deposits (Fig. 1b). They are formed essentially by sandstone and marl deposits. Fig. 3b, c show the thicknesses of outcrop units, deduced from the geological maps (1/50000) of Nabeul, Grombalia, Menzel Bou Zalfa, and la Goulette. The Grombalia groundwater system is composed by shallow (SGW) and deep groundwater (DGW). They are separated by a 15 m-thick clay layer (Ben Moussa et al. 2009). The SGW is formed by the Quaternary sediment that is composed by alluvium, sand, and sandy clay, with 50-m depth. The DGW is composed by Mio-Plio-Quaternary (Segui Formation), Miocene (Beglia Formation), and Oligocene series. The Mio-Plio-Quaternary is characterized by geologic and geometric complexities and it is formed by intercalation of sand, sandy clay, and clay deposits.

The study area is characterized by a Mediterranean semi-arid climate with an average annual rainfall of 486 mm (calculated during the 1898–2014 period in Grombalia station). Rainfall is characterized by a high space-time variability; it fluctuates between 192 and 1063 mm in 1987/1988 and 2003/2004, respectively. The main rainfall (60%) occurs



aQ:Recent and current alluvial; dQ: Coastal dune; spQ: Sebkhats basins; ^{m}Q: Middle and upper Pleistocene; ^{c}Q: Middle and upper continental Pleistocene; P1:marine Pliocene; M-P1:Mio-Pliocene continental; ^{a}M$_3$: Upper Miocene; M$_{1\text{-}2}$: Lower and Middle Miocene; M$_1$: Aquitanian; O:Oligocene; ^{a}E$_{2\text{-}3}$:Lutetian-Priabonian; ^{g}E$_1$:Ypresian; Cm$_2$-p:upper Maastrichtian-Paleocene; ^{cd}C$_2$:Senonian to lower Eocene; ^{c}C$_2$:lower Senonian; ^{a}C$_2$:Vraconian to Turonian; ^{s}C$_1$:lower Cretaceous; T:Triassic.

Fig. 1 Grombalia basin location. **a** Geographical location, **b** geological map of the study area (Ben Haj Ali et al. 1985), and **c** water sample locations of Grombalia basin between November and March. The mean annual temperature is 17 °C (Charfi et al. 2013).

Studied aquifer is one of the typical examples of coastal aquifers in semi-arid region that have been intensively overused during the last few years (Gaaloul et al. 2014; Lachaal et al. 2016). The general piezometric level dropped and the water quality is relatively deteriorated. In this context, and in addition to the local surface water, the government have added the use of surface water transferred from the North-west Tunisia (Medjerda and Ichkeul basins), since 1984. These resources are used for irrigation and drinking water purposes.

Materials and method

Groundwater samples were collected in April 2015 from 39 wells (37 from SGW and two from DGW) (Fig. 1c). In addition, two samples were collected from El Bay River and two samples from the North-west surface water. The water sampling was concomitant with the piezometric measurements. Water was stored in clean polyethylene bottles. Temperature (*T*), electrical conductivity (EC), and pH were measured in situ. SO_4^{2-} concentration was determined using the gravimetric method. Cl^- was analyzed using titration (Mohr method). HCO_3^- and CO_3^{2-} were determined by titration with sulfuric acid. NO_3^- was measured by spectrophotometer. Cations Ca^{2+}, Na^+, Mg^{2+}, and K^+ were analyzed by atomic absorption spectrometer.

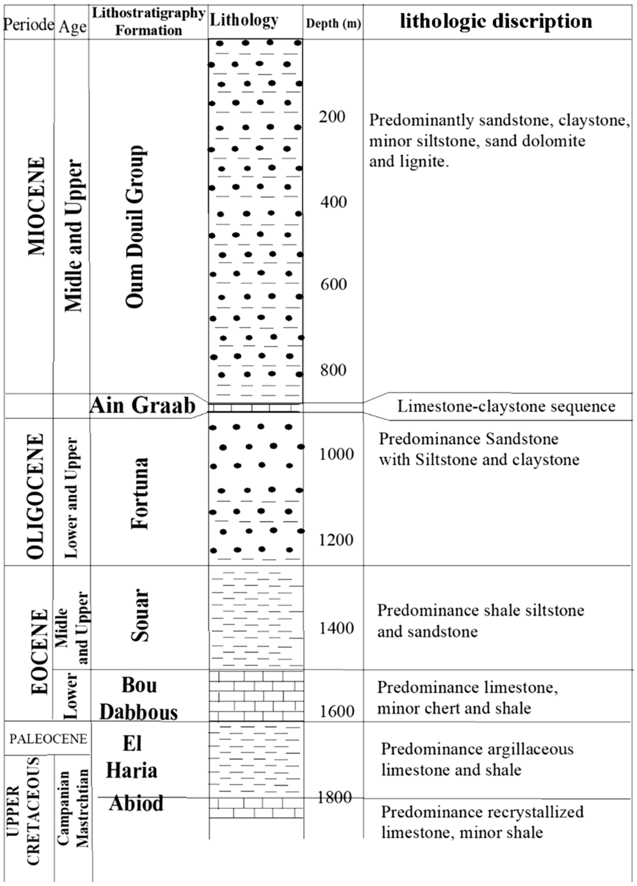


Fig. 2 Lithostratigraphic column of Belli-3 petroleum well (BEL-3)

Piezometric data were collected from 73 monitoring wells in April 2015 (Table 1). They were used to draw the contour line map (Fig. 4). In addition, historical data spanning over the 1973–2014 period were used. These data were provided by the local water authority of regional commissioner of agricultural development of Nabeul (CRDA Nabeul). It consists of monthly water-level measurements in 35 monitoring wells covering the aquifer area (Fig. 4).

A complementary set of hydrological data was collected also from CRDA Nabeul. It covers the monthly precipitation data of the Grombalia station covering the period 1973–2014, the deep Grombalia aquifer abstraction of 1989–2013 period, the North-west water resources used in irrigation, and the artificial recharge volume of Grombalia aquifer during the 1992–2014 period. The piezometric and water depth maps were drawn using the kriging interpolation method.

Agriculture irrigation, water supply, and anthropogenic activities

Agriculture irrigation

Grombalia region economy is based on agriculture. Indeed, the agricultural cropping is principally dominated by citrus, vines, grain, and vegetable crops. In Beni Khalled and Menzel Bouzelfa regions, the essential of the irrigated area is planted by citrus (77.26% of the irrigated area). Nabeul district production of citrus represents about 255,000 tons in 2014 which represent 78% of the total national production of citrus (Lakani 2014). These cultures are considered as the most consumers of water. Its exportation to the Mediterranean and European countries provides 19.9 Million Tunisian Dinars which is very benefic to the economic balance of Tunisia (Onagri 2015).

The total area of public irrigated perimeters in the Nabeul district is approximately about 268 km^2 in 2014. It represents about 54.5% of total irrigated area in the prefecture which are estimated to 492 km^2 ha. The public irrigated perimeters areas were divided into two domains:

- The safeguard irrigated perimeters (SIP): it is characterized by intensive irrigation activities, because of its social and economic importance. This area is cropped with citrus, which need important water quantity with good quality. The SIP is equipped with public water supply system that contributes to filling a percentage of water needs for irrigation. The water irrigation is supplied from the North-

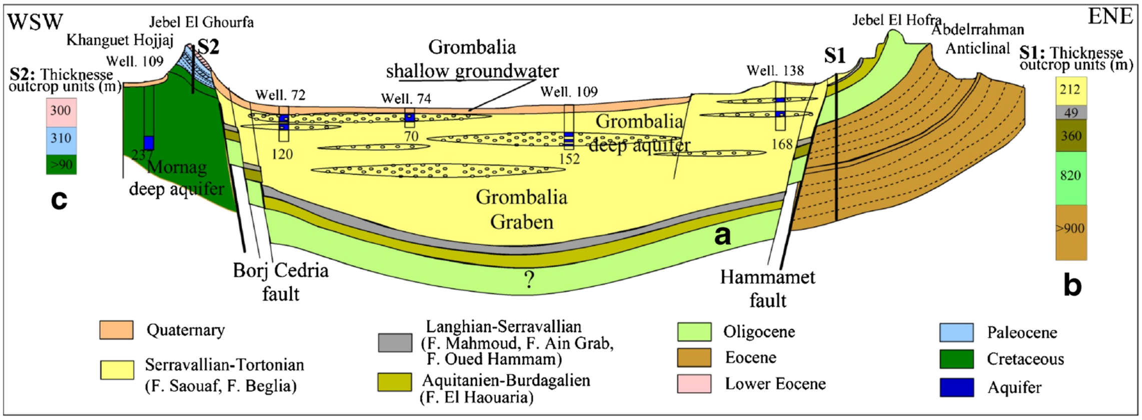


Fig. 3 Schematic and conceptual model of Grombalia aquifer system A-A′. **a** Schematic aquifer system, showing that the aquifer is limited to the East-north-east by the Jebel El Hofra and to the West-south-west by Jebel Ghourfa, Jebel Djemaa, Jebel Messaoud, and Mornag hydrogeologic basin. **b**, **c** Thicknesses of outcrop units, deduced from the geological maps (1/50000) of Nabeul, Grombalia, Menzel Bou Zalfa, and la Goulette

west surface water, while the farmers provide the rest of irrigation water needs through the DGW pumping. These areas cover about 137.45 km^2 from which 121.45 km^2 are citrus backups perimeters in Beni Khalled, Menzel Bouzelfa, Soliman, Grombalia, and Bou Argoub regions while 160 km^2 are related to Korba-Mida backups (KMT) which are outside of the study area.

- The public irrigated perimeters (PuIP): covers approximately 13,000 ha (130 km^2) cultivated mainly by vegetables, strawberry with fruit trees, and field crops principally in the Grombalia region. The water needs for PuIP are supplied from the North-west surface water.
- The private irrigated perimeters (PrIP): are irrigated by SGW and DGW. It covers the upstream aquifer, and the aquifer boundaries in East and West sides with a total area of 7083 ha (70.83 km^2), about 21.1% of groundwater abstraction area.

Water resources management in the Grombalia basin

Due to the growth of water demand, especially for the agricultural activities, the water needs is supplied by four origins: local surface water, SGW, DGW, and transferred water from the North-west of Tunisia.

Grombalia shallow aquifer system has been considered as an important water resource for long period, because of its good quality and its availability, especially, for the local population and development of agricultural and industrial sectors. In order to meet the growing water needs, aquifers are the main source for irrigation water supply. Since 1959, the SGW is overexploited, i.e., the intensive groundwater exploitation to meet the water needs has caused a decrease in groundwater levels and degradation of the water quality.

In order to increase the local available water resources, the government implemented surface water dams. The principal reservoirs were the Bezigh dam built in 1954, with 6.5 hm^{3}year^{-1} capacities and it is located in the upstream of Menzel Bou Zelfa city, the Tahouna dam built in 1967, with 0.96 hm^{3}year^{-1} capacities, and El Masri dam edified in 1968, with 6.9 hm^{3}year^{-1} of capacity and it is located in upstream of Bou Argoub city.

Since 1973, groundwater artificial recharge schemes were conducted in order to increase the piezometric level in depleted areas located in Bou Argoub and in the eastern side of the aquifer.

Since 1984, and because of insufficiency of local water resources issues for irrigation water needs, especially, for circus crops, the Ministry of Agriculture has transferred water from the North-west Tunisia (Medjerda and Ichkeul basins).

The Grombalia water resources management is very complex. In fact, the water needs are supplied from the SGW and DGW, local dams and from the North-west surface water. In Grombalia basin, different water origins are used in agriculture. In addition to the use of surface water of Medjerda-Cap Bon canal, the farmers are also using SGW and DGW.

Anthropogenic activities

The groundwater pollution is caused by two types of anthropogenic activities. The first one is the discharge of urban and industrial wastewater and solid waste into rivers. The second type is related to the agricultural pollution caused by cumulative use of fertilizers, pesticides, and other chemical products in irrigated areas.

The industrial activities in these regions are located in Grombalia, Soliman, Bou Argoub, and Menzel Bouzelfa. They are essentially formed by:

(i) Chemical industries: using organic and inorganic chemicals during processing and washing production processes;

Table 1 In situ measurements and geochemical data of water resources in Grombalia basin

Samples number	Water type	T °C	pH	TDS (mg L^{-1})	Ca (mg L^{-1})	Mg (mg L^{-1})	Na (mg L^{-1})	K (mg L^{-1})	HCO_3 (mg L^{-1})	Cl (mg L^{-1})	SO_4 (mg L^{-1})	NO_3 (mg L^{-1})	BI (%)
1	SGW	18.9	7.71	4.6	360.72	214.8	781.7	34.9	1330	958.5	619.8	292.3	3
3	SGW	19.9	7.49	3.7	296.5	220.8	549.2	15.8	1295	958.5	204.6	159.8	2
4	SGW	20.8	7.69	5.3	545	318.7	801.7	15.9	1501	1552.5	455.8	152.1	5
7	SGW	19.4	7.71	4.5	482	248.7	700	14.7	1285	1290.	330.3	198.6	5
10	SGW	26.2	7.6	4.7	340.6	222.3	672.2	10	1330	1065.5	316.	35.6	5
11	SGW	18.2	7.81	4.0	348.6	250.32	644.5	37	1242.5	1052	419	119	5
13	SGW	19.9	7.87	4.1	348.6	195.3	590	13.8	1102.5	986	328.7	102	4
15	SGW	19	7.71	3.7	501	217.25	718	11.7	1295	1321	325.2	129.5	5
16	SGW	18.4	7.71	4.5	541	234.7	645.2	4.41	1190	1701	560.11	212.3	−5
17	SGW	19.3	7.73	5.1	440.8	262.2	625.5	12.8	1263	1203	378.14	138.6	5
23	SGW	19.3	8.17	4.3	641.2	367.1	1123	88.01	945	2804.5	358.9	88.23	5
24	SGW	18	7.97	6.4	525	213.7	643.25	58.7	980	1633	271.64	68.68	3
25	SGW	21.2	7.91	4.4	416.8	238.2	832	34.9	962.5	1526.5	431.6	130.86	5
26	SGW	21.5	7.87	4.6	401	209.7	812	20.31	950	1430	363.79	149.2	5
28	SGW	18.4	7.97	4.3	424.8	198.7	802	17.1	980	1286	523	140.1	5
29	SGW	18.1	7.93	4.4	462	250.2	1065	44.5	1050	1392	1040.8	280.2	5
30	SGW	20.6	7.96	5.6	402	228.79	1253	14.87	927.5	1810.5	896.51	0.22	5
33	SGW	21.2	7.99	5.5	292.5	234.3	503	16.3	1067.5	923	233.8	168.6	5
34	SGW	20.6	7.62	3.4	392.7	211.8	343.5	3.61	1207.5	852	230.5	147.8	1
36	SGW	20.8	7.83	3.4	288.57	160.35	497.75	6.98	840	745.5	440.61	43.19	5
44	SGW	20.3	8.13	3.0	501	204.7	653.25	8.4	1155	1023	660.2	115.4	5
46	SGW	20.1	8	4.3	360.7	184.8	574.2	11.7	1312.5	781	601.1	68.7	1
47	SGW	18.6	7.88	3.9	336.6	135.8	399	7.3	1120	674.5	348.5	2.3	1
54	SGW	20.4	7.87	3.0	400.8	172.3	616.75	9.11	1225	905	397.49	88.84	5
56	SGW	20.2	8	3.8	280	110	476.75	52.88	571.5	786	399.19	69.4	5
58	SGW	20.6	8.13	2.7	504	194	905	14.18	472.15	1165	1381.1	210.42	5
59	SGW	19.4	7.9	4.8	532	204.5	923	73	875	1484.5	760.46	291.2	5
61	SGW	21.7	8.4	5.1	316	151	585.2	16.5	1190	786	261.1	127.1	5
64	SGW	21.3	8.2	3.4	296	181	539	9.72	875	921	321.1	91.9	5
66	SGW	24.2	8.1	3.2	364	184	119.2	9.72	889	745.5	341.7	4.9	−5
68	SGW	21.8	7.63	2.7	716	555	133.5	24.8	1260	1633	787.6	153.6	1
69	SGW	23	7.7	5.3	760	642.5	120	28.8	1260	1917	492.6	328.7	3
70	SGW	23.3	7.6	5.5	712	579.5	120	14.8	980	1668.5	474	343.4	6
73	SGW	23.6	8.04	4.9	204	421.5	129.2	12.9	1260	1170.5	1370.8	9.161	−5
74	SGW	24.0	8.4	4.6	468	210.5	135	11.13	1120	781	200.4	318.2	−3
76	SGW	24.3	7.72	3.2	180	412.5	712	14.85	1225	1032	845.3	26.8	5
77	SGW	19.4	7.95	4.4	300	197.5	416	24.4	1155	545	500.8	32.4	5
6	DGW	21.2	7.85	3.2	344.6	231.3	485.2	0.17	686	1203	299	7.3	5
35	DGW	21.4	7.95	3.3	224.4	121.3	390.7	6.24	1032.5	521	120	38.4	5
42	SW	18.8	8.9	2.5	136.2	78.4	403	8.9	507.5	491	297.3	15.9	4
67	SW	21.1	8.12	1.9	140	87.5	230	12.8	525	461.5	252.9	6.4	−5

SGW, shallow groundwater; DGW, deep groundwater; SW, North-East surface water

(ii) Paper industries: the paper industry use products that are generally toxic containing MES of organic materials, phenolic compounds, organochlorine compounds, and nutrients;
(iii) Agricultural and food industries: characterized by high levels of dissolved organic matter and suspended solids.

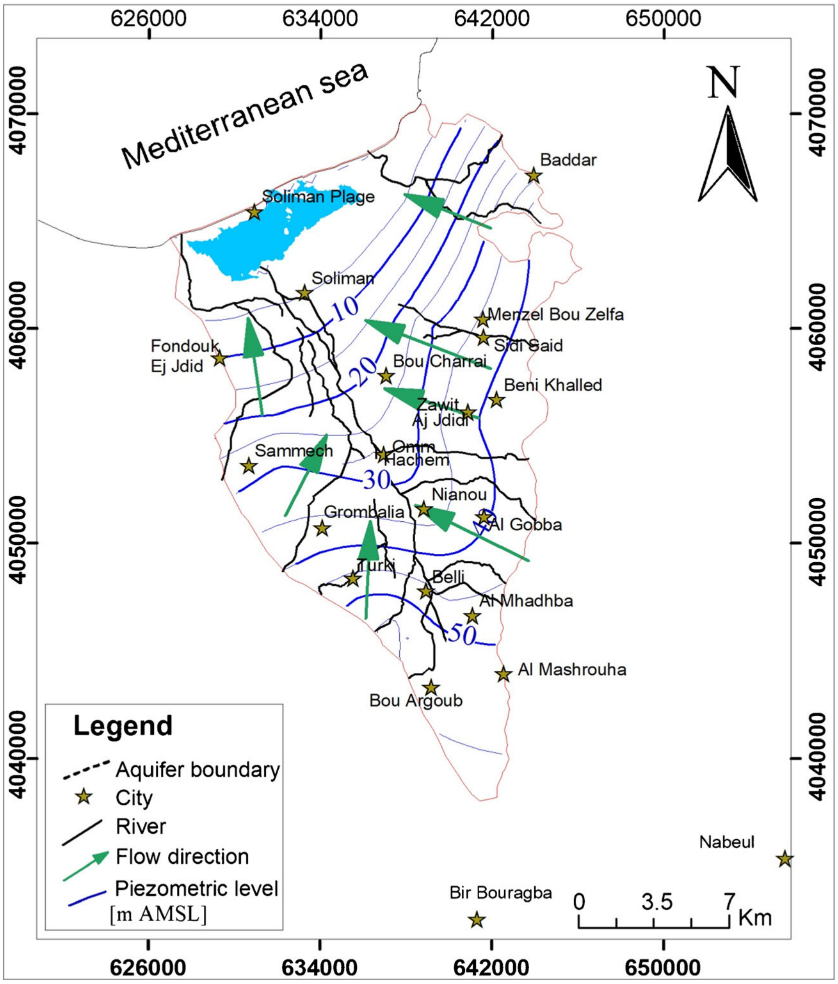
Results and discussion

Groundwater piezometric rise

We have drawn the piezometric map of April 2015, using the interpolation of 77 observation wells (Fig. 4). The measured water level is varying between 1.9 m near the Sebkhat El Meleh and 60 m in upstream in Bou Argoub region. The piezometric map shows a general water flow from the South-east to North-west. The geometry of piezometric contours is concave, its curve is opening downstream, and the flow lines converge to the favorite drainage axis coinciding with el Bey River. The piezometric lines form a parabolic form. The hydraulic gradient is varying from 3‰ in the center to 5‰ in the East side. Sebkhat El Melah and the sea represent the groundwater outlet.

Grombalia shallow groundwater is characterized by an increase in water level in the central part (Fig. 5). Since 1992, the groundwater rose up to 2 m from the ground surface, and creating many agricultural, environmental, and economical problems. These problems are linked to suffocation of plants,

Fig. 4 Piezometric map of shallow Grombalia aquifer in April 2015



trees, and crops. The field observations and discussions with farmers during 2013–2015 period showed that in recent years, the groundwater rise exceeded the root zone level and asphyxiated some fields of lemon trees. The water-level depth map shows the presence of groundwater rise in the central part, between Beni Khalled, Menzel Bou Zelfa, Grombalia, and Soliman towns, and it is concentrated in Zaouiet Jedidi, Omm Hashem, Beni Khalled, Henchir Bou Charaya, and Soliman regions**.**

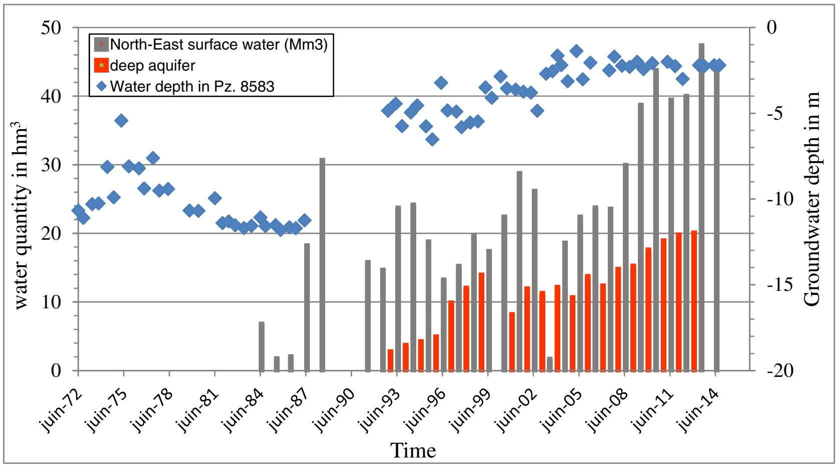
Figure 6 shows a good correlation between the groundwater rise and the use of North-west water resources and DGW extraction. The increase of water level is correlated to the increase of (i) the DGW pumping and (ii) the use of North-west water for irrigation. The excess irrigation return-flow is increasing the natural groundwater direct recharge component. In fact, in addition to the local water resources, the government has added since 1984 the use of water resources from the North-west Tunisia (Medjerda and Ichkeul basins). The used North-west water resources in agriculture with an average of 5000 m^3 ha^{-1} $year^{-1}$ enhanced the infiltration and the water level rise by irrigation returns.

The piezometric rise area is characterized by the intense irrigation activities especially for the SIP which needs a good water quality. The water need is supplied from the North-west surface water and from the DGW. The farmers use DGW in citrus irrigation because of good quality of the irrigation water, especially in Beni Khalled, Menzel Bouzelfa, and Soliman zones. These resources contributed also to the groundwater recharge by irrigation returns.

Groundwater physicochemical parameters

Temperature of SGW varies between 18 and 26.2 °C (Table 1). The pH values are varying between 7.49 and 8.45, with an average of 7.89, indicating an alkaline composition (Table 1). The pH wide variation is caused by two origins. The first is the impact of the anthropogenic pollution from the infiltration of domestic and industrial wastewater, produced especially in Soliman and Grombalia regions. The second is the return flow from irrigation water characterizing Menzel Bouzelfa, Beni Khaled, and Bou Argoub regions (Ben Moussa et al. 2008; Lachaal et al. 2016).

Fig. 5 Correlation between shallow groundwater piezometric unregistered in 8583/2 piezometer with North-East water used in irrigation and deep aquifer exploitation in Grombalia basin



The EC and the total dissolved solids (TDS) of SGW samples are ranging from 2.2 to 6.7 mS cm^{-1} and from 2.6 to 5.6 g L^{-1}, respectively (Table 1). The higher values characterize the wells situated in central and downstream of the aquifer, especially, in the region between Nianou and Turki towns. However, wells in the upstream (southern parts) are marked by low values, especially, in Grombalia, Beni Khaled, and Bou Argoub regions. Wide variation of EC and TDS in SGW parameters is caused by both natural and anthropogenic mineralization processes. The natural processes interaction water-rock and mixing processes related to the return flow of irrigation. The anthropogenic processes are the pollution by industrial wastewater.

Fig. 6 Spatial distribution of the salinity in the Grombalia shallow groundwater in April 2015

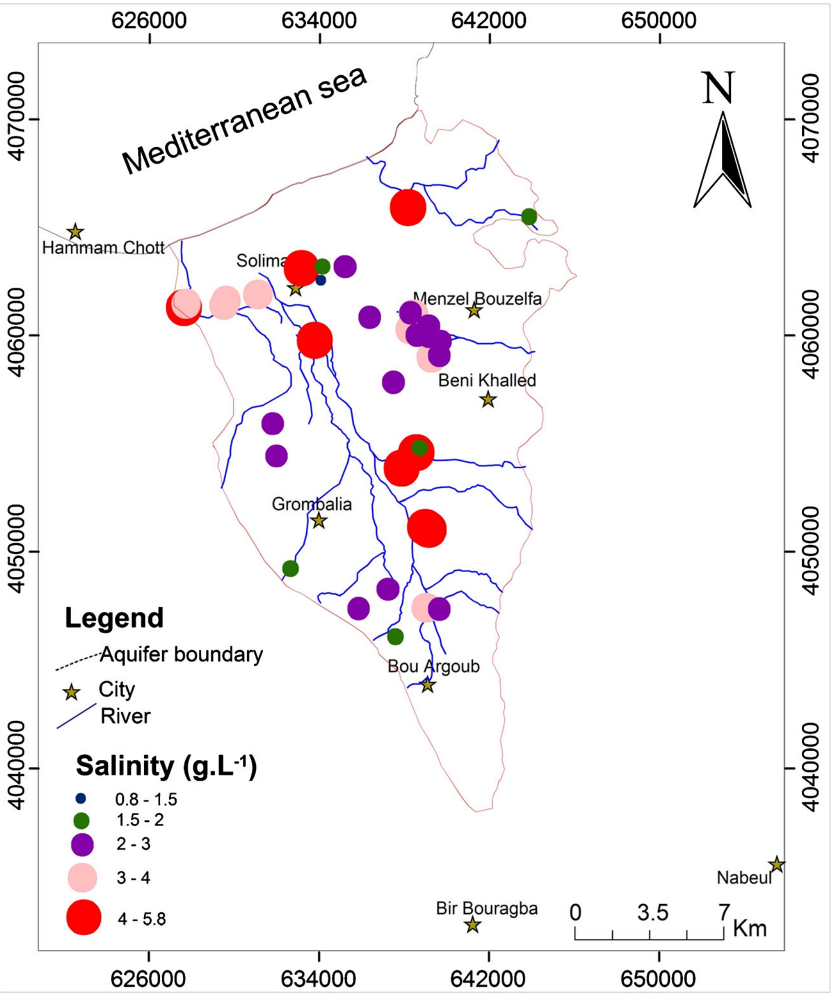
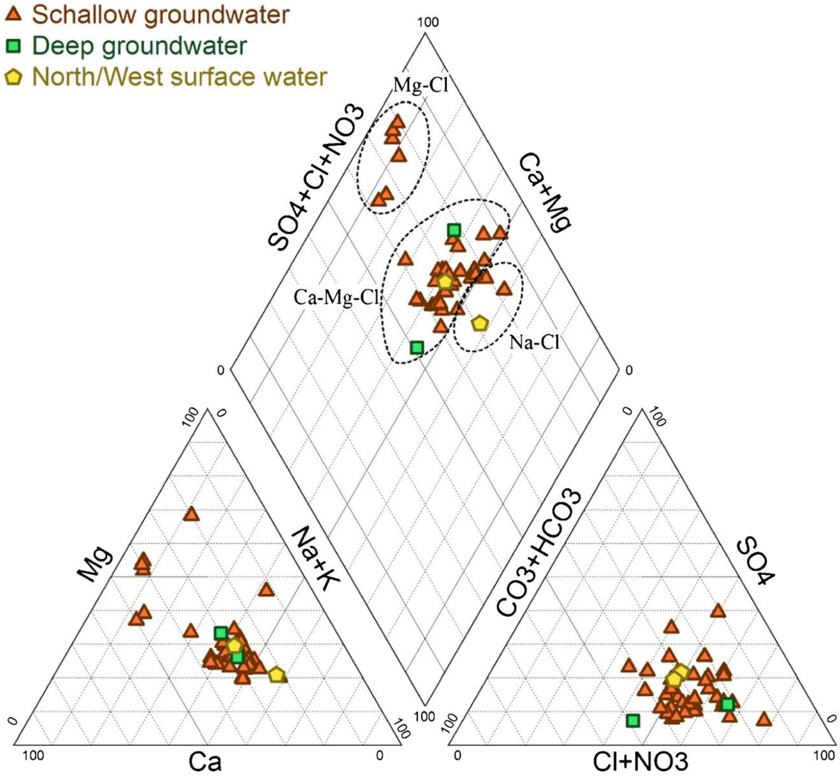


Fig. 7 Piper diagram of Grombalia water samples



DGW was slightly alkaline with pH of 7.85 and 7.95. The EC are 1.63 and 3.38 mS cm^{-1}, the TDS are 2.4 and 3.2 g L^{-1}.

The SW is characterized by good quality compared to groundwater samples. SW is alkaline composition with pH varying between 8.12 and 8.9, and a low salinity varying between 1.7 and 1.9 g L^{-1}.

Groundwater mineralization processes

Groundwater mineralization

The salinity values of SGW samples range from 1.54 to 5.81 g L^{-1} (Fig. 6). The salinity distribution revealed the presence of salinization gradient from East to West, corresponding to the main groundwater flow direction. The lowest salinity was observed in the upstream part, which varied between 1.5 and 2.5 g L^{-1} reveal the influence of recharge processes, it is characterized by the presence of high level of limestone. However, a high salinity was observed in two distinct regions: The first one is located in the northwestern of the basin close to Soltane River (5.81 g L^{-1}). The second is situated in the central part of Grombalia basin between the Nianou and Turki towns (4.6 g L^{-1}). The higher salinity values observed in the first region are attributed to high contamination by industrial wastewater, especially, in Soliman region. The high salinity is observed also in the central aquifer which coincides with the piezometric rise and groundwater management problem. The high salinity can be explained by the leaching of soil with high nitrate concentration, and the lithology composition of saturated and unsaturated layers. The water flow in this region is lower and the residence time is higher, causing enrichment with Na-Cl (Mlayah et al. 2017).

Triassic outcrops, around the Jebel Ressas diaper, are located in the West side of the study area. Oued Al Bakbaka flows from the west flank of Jebel Ghourfa to the West along the Khanget Hojjaj syncline. It is located in the Mornag basin, it is an affluent of River Meliane, and it is outside of Grombalia hydrogeological basin. However, oued Bou Abid flows from the East flank of Jebel Ghourfa to the East and it represent an affluent of El Bay River. Oued Al Bakbaka and oued Bou Abid basins are separated by Jebel Ghourfa. Jebel Ghourfa, Jebel Djemaa, and Jebel Messaoud are the west limit of Grombalia basin. As conclusion, the dissolution of Triassic evaporates that are rich in salt and gypsum minerals do not contribute in Grombalia groundwater mineralization. Triassic outcrops can have an impact on the mineralization of Mornag aquifer system. However, halite and gypsum dissolution are the dominant water-rock processes controlling the water salinity. These minerals are abundant in the Miocene series (Mannaï-Tayech 2009; Ben Moussa et al. 2009; Lachaal et al. 2010).

Hydrogeochemical water types

In order to distinguish different water facies, major ions of SGW, DGW, and SW were plotted on the Piper diagram

Table 2 Saturation indices of water resources in Grombalia basin

Samples number	Water type	Is calcite	Is aragonite	Is dolomite	Is gypsum	Is anhydrite	Is halite
1	SGW	1.45	1.3	2.96	−0.71	−0.95	−4.81
3	SGW	1.21	1.06	2.58	−1.21	−1.45	−4.96
4	SGW	1.63	1.48	3.33	−0.75	−0.99	−4.62
7	SGW	1.55	1.4	3.09	−0.86	−1.1	−4.79
10	SGW	1.44	1.3	3.08	−1.02	−1.24	−4.84
11	SGW	1.51	1.37	3.16	−0.88	−1.12	−4.87
13	SGW	1.57	1.42	3.18	−0.94	−1.18	−4.97
15	SGW	1.59	1.45	3.11	−0.86	−1.1	−4.76
16	SGW	1.55	1.4	3.01	−0.63	−0.87	−4.56
17	SGW	1.51	1.36	3.08	−0.84	−1.08	−4.86
23	SGW	1.92	1.77	3.89	−0.85	−1.09	−4.14
24	SGW	1.73	1.58	3.34	−0.91	−1.15	−4.67
25	SGW	1.59	1.45	3.26	−0.84	−1.07	−4.5
26	SGW	1.54	1.39	3.08	−0.87	−1.1	−4.55
28	SGW	1.63	1.49	3.21	−0.77	−1.01	−4.55
29	SGW	1.65	1.5	3.25	−0.42	−0.66	−4.46
30	SGW	1.58	1.43	3.19	−0.57	−0.8	−4.27
33	SGW	1.62	1.47	3.46	−1.17	−1.4	−5.01
34	SGW	1.44	1.29	2.91	−1.05	−1.28	−5.21
36	SGW	1.37	1.22	2.78	−0.85	−1.08	−5.1
44	SGW	1.94	1.79	3.79	−0.56	−0.8	−4.91
46	SGW	0.85	0.71	1.76	−0.7	−0.92	−5.04
47	SGW	1.58	1.43	3.04	−0.87	−1.11	−5.23
54	SGW	1.67	1.52	3.28	−0.81	−1.05	−5
56	SGW	1.43	1.29	2.81	−0.87	−1.09	−5.21
58	SGW	1.52	1.37	2.92	−0.28	−0.51	−4.67
59	SGW	1.65	1.5	3.17	−0.52	−0.76	−4.47
61	SGW	2.11	1.97	4.24	−1.07	−1.3	−5.08
64	SGW	0.64	0.5	1.43	−1	−1.22	−5.08
66	SGW	1.85	1.7	3.75	−0.88	−1.1	−5.72
68	SGW	1.61	1.47	3.44	−0.49	−0.72	−5.38
69	SGW	1.69	1.54	3.61	−0.69	−0.92	−5.36
70	SGW	1.49	1.34	3.2	−0.7	−0.94	−5.42
73	SGW	1.47	1.32	3.56	−0.67	−0.91	−5.52
74	SGW	2.2	2.06	4.39	−1.05	−1.28	−5.65
76	SGW	1.15	1	2.97	−0.93	−1.16	−4.94
77	SGW	1.61	1.46	3.31	−0.79	−1.03	−5.65
6	DGW	1.09	0.95	2.32	−0.98	−1.21	−4.95
35	DGW	1.53	1.39	3.11	−1.43	−1.66	−5.47
42	SW	1.82	1.67	3.69	−1.2	−1.44	−5.32
67	SW	1.24	1.1	2.59	−1.21	−1.44	−5.82

SGW, shallow groundwater; DGW, deep groundwater; SW, North-east surface water

(Fig. 7) (Piper 1944). Nitrate concentration was taken into account because of its abundance in the SGW. According to the Piper diagram (Fig. 7), SGW present tree water types: Na-Cl facies, mixed Ca-Mg-Cl facies, and Mg-Cl facies.

- Na-Cl water type characterizes SGW and it was found in wells 25, 26, 28, 29, and 30 located mainly along the coastal region and near Sebkhet El Malah, which corresponds to a discharge zone.
- A mixed Ca-Mg-Cl water type characterizes SGW and DGW and it was identified in the artificial recharge zone, as well as, in the south part of the aquifer. This facies shows no dominant cation, which signifies the combination of the two natural processes: dissolution of evaporatic minerals and cation exchange or mixing process.
- Mg-Cl water type, which characterizes the wells 66, 68, 69, 70, 73, and 74, situated in the central part of the aquifer. Cl and Mg were the dominant ions. This water type can indicate the influence of the both, natural process probably the dissolution of evaporatic minerals (Halite) and the ion exchange (Mlayah et al. 2017).

Saturation indices

The saturation indices (SI) of minerals such as calcite, dolomite, aragonite, halite, gypsum, and anhydrite were calculated using the tri-linear Piper diagram. The results are presented in Table 2. SI less than zero indicate that the groundwater is undersaturated with mineral. SI more than zero, specifies that groundwater being supersaturated with mineral and incapable of dissolving more of the mineral (Benony 2006).

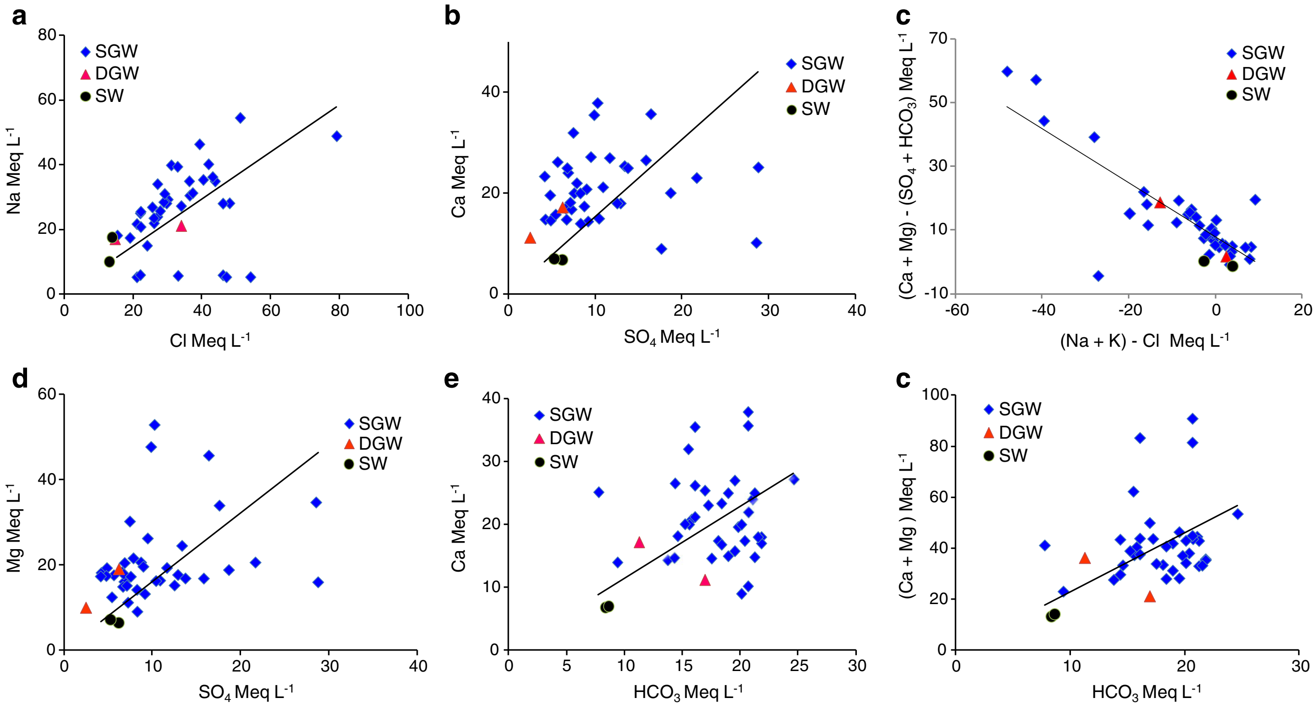


Fig. 8 Major elements relationships of water resources in Grombalia basin

Most of the water samples (85%) are undersaturated with gypsum and anhydrite. The SI is varying from − 1.43 to − 0.28 and from − 1.66 to − 0.51 for gypsum and anhydrite, respectively (Table 2). Sulfate dissolution may occur for the majority of groundwater samples. The groundwater and surface water samples are undersaturation state with halite, the SI varying from − 5.82 to − 4.14, indicating possible dissolution of these evaporatic minerals. The water mineralization origin is the water-rock interaction in three zones: recharge area, vadose zone, and saturated layer.

Origins of major ions

Figure 8a shows significant correlation between water samples and halite dissolution line, confirming that halite dissolution is an important mineralization origin. The linear relationship between Na and Cl concentration of most groundwater samples present an excess of Na. As a result, Grombalia aquifer was influenced by halite dissolution and ion exchange mechanism between Ca/Mg in groundwater and Na-clay minerals. However, some saline groundwater samples (66, 68, 69, 70, 73, and 74) were plotted away from halite dissolution line suggesting that the reduction of Na concentration may be due to reverse ion exchange reaction. Sodium and chloride were also significantly correlated with TDS, $r^2 = 0.8$ and $r^2 = 0.9$ for sodium and chloride, respectively.

The Ca/SO_4 diagram shows that most groundwater samples are an excess of calcium (Fig. 8b). It can be attributed to the release of Ca cation through the dissolution of gypsum and anhydrite. Another process that can account for the excess of Ca is the reverse cation exchange, occurring between groundwater and Ca-clay minerals, such as Illite, Kaolinite, Sepiolite, and Montmorillonite, which are relatively abundant in the Mio-Plio-Quaternary sediment. In fact, reverse cation exchange process is confirmed through the plot of (Na + K–Cl) versus [(Ca + Mg)–(HCO_3 + SO_4)], in which the two members vary in inverse proportions (especially for 95% of wells located in Mio-Plio-Quaternary sediment) (Fig. 8c). Mg/SO_4 diagram (Fig. 8d) shows a moderate correlation suggesting a common origin, possibly related to Epsomite dissolution ($MgSO_4 7H_2O$) (Adams et al. 2001) or the contribution of $MgSO_4$-fertilizers (Gi-Tak et al. 2004) commonly used in agriculture areas especially in Menzel Bouzelfa and Beni

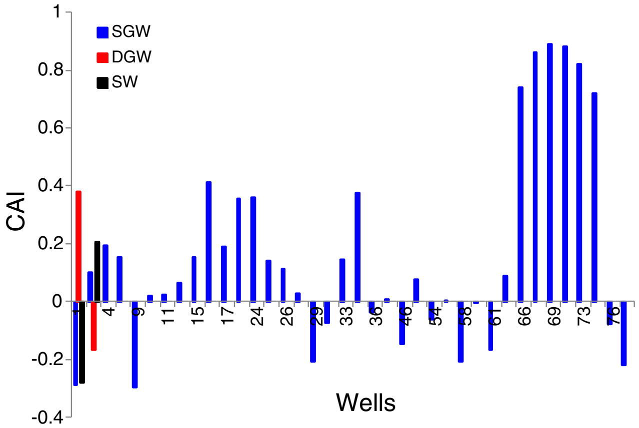


Fig. 9 Chloro Alkaline Index (CAI) of Grombalia groundwater and North-east surface water

Table 3 Correlation matrix of chemical parameters in Grombalia groundwater aquifer

Variables	Ca	Mg	Na	K	HCO_3	Cl	SO4	NO_3	salinity	T°C	pH
Ca	1	0.644	0.095	0.338	0.259	0.761	0.141	0.644	0.746	− 0.126	− 0.346
Mg	0.644	1	− 0.231	0.152	0.375	0.618	0.324	0.420	0.640	0.139	−0.437
Na	0.095	− 0.231	1	0.407	− 0.012	0.434	0.256	0.036	0.525	− 0.328	− 0.039
K	0.338	0.152	0.407	1	− 0.123	0.542	0.114	0.241	0.458	− 0.337	0.052
HCO_3	0.259	0.375	− 0.012	− 0.123	1	0.148	− 0.045	0.226	0.385	0.009	− 0.548
Cl	0.761	0.618	0.434	0.542	0.148	1	0.244	0.351	0.885	− 0.162	− 0.281
SO_4	0.141	0.324	0.256	0.114	− 0.045	0.244	1	0.120	0.513	− 0.086	− 0.025
NO_3	0.644	0.420	0.036	0.241	0.226	0.351	0.120	1	0.490	− 0.197	− 0.230
salinity	0.746	0.640	0.525	0.458	0.385	0.885	0.513	0.490	**1**	− 0.207	− 0.392
T°C	− 0.126	0.139	− 0.328	− 0.337	0.009	− 0.162	− 0.086	− 0.197	− 0.207	1	− 0.046
pH	− 0.346	− 0.437	− 0.039	0.052	− 0.548	− 0.281	− 0.025	− 0.230	− 0.392	− 0.046	1

Khalled regions (Ben Moussa et al. 2008; Lachaal et al. 2016). SO_4 is attributed to the (Ca SO_4, $4H_2O$) dissolution witch abundant in the Mio-plio-quaternary sediments.

The Ca/HCO_3 and (Ca + Mg)/HCO_3 diagrams show a poor correlation between this major components (Fig. 8e, f). In their present state, sampled waters cannot dissolve calcite and dolomite, as they are oversaturated or close to equilibrium with respect to these minerals. Thus, the relative excess of Ca and Mg appear to be mainly derived from the dissolution of anhydrite and gypsum and ionic exchange process.

A cross-plot of HCO_3^- with TDS shows no linear relationship between them in more loaded groundwater samples indicating the poor participation of this element in the natural mineralization process.

Ion exchange reactions

The water geochemistry was also analyzed using the indices of Base Exchange, called the Chloro Alkaline Index CAI (Garcia et al. 2004). These indices are calculated using the following equation Eq. (1):

$$\mathrm{CAI} = [\mathrm{Cl}-(\mathrm{Na}+\mathrm{K})]/\mathrm{Cl} \qquad (1)$$

If there is exchange between Na or K from water with Ca or Mg in the aquifer minerals, the index is positive, highlighting reverse ion exchange reactions. If the later values are negative, then there is direct ion exchange and therefore, there are exchange reactions between Ca or Mg in water with Na or K in the aquifer materials (Hamzaoui-Azaza et al. 2011; Mlayah et al. 2017).

The most of SGW samples are characterized by positive index, especially, for wells between Nianou and Turki towns (66, 68, 69, 70, and 74 wells) (Fig. 9). The CAI is varying between 0.74 and 0.89 indicating the reversion exchange reaction between Na or K from water with Ca or Mg in the aquifer minerals. This result confirms that this process control the Mg-Cl facies, which characterizes the wells of aquifer central part. Furthermore, the contribution of this process is confirmed by the diagram of ((Na + K–Cl)/[(Ca + Mg) − (HCO_3 + SO_4)]) (Mc Lean et al. 2000; Garcia et al. 2001; Ben Moussa et al. 2012).

Table 4 Statistical correlations between variables and factors for the hydrogeochemical parameters of water resources in Grombalia basin

Factors	F1	F2	F3	F4	F5	F6	F7	F8	F9	F10	F11
Ca	0.848	− 0.137	− 0.148	− 0.301	0.040	0.115	− 0.069	− 0.214	0.281	− 0.068	0.000
Mg	0.727	− 0.474	− 0.348	0.039	− 0.001	− 0.261	0.036	− 0.019	− 0.202	− 0.115	0.000
Na	0.367	0.677	0.388	0.333	0.167	0.317	− 0.010	− 0.030	− 0.082	− 0.097	0.000
K	0.502	0.574	− 0.066	− 0.264	0.269	− 0.246	0.116	0.439	0.072	− 0.008	0.000
HCO3	0.395	− 0.552	*0.560*	0.122	− 0.068	− 0.056	0.445	0.033	0.056	0.017	0.000
Cl	0.881	0.174	− 0.112	0.000	0.320	− 0.059	− 0.030	− 0.220	− 0.089	0.132	0.000
SO4	0.393	0.209	− 0.364	*0.651*	− 0.467	− 0.041	− 0.013	0.123	0.104	0.027	0.000
NO3	0.621	− 0.123	− 0.058	− 0.460	− 0.392	0.415	− 0.005	0.208	− 0.116	0.033	0.000
Salinity	0.963	0.110	0.018	0.212	0.016	0.054	0.082	− 0.069	− 0.014	0.024	0.000
T °C	− 0.231	*− 0.540*	− 0.406	0.288	*0.489*	0.354	0.090	0.185	0.033	0.007	0.000
pH	− 0.478	0.518	− 0.449	− 0.172	− 0.085	0.081	0.464	− 0.205	− 0.024	− 0.014	0.000

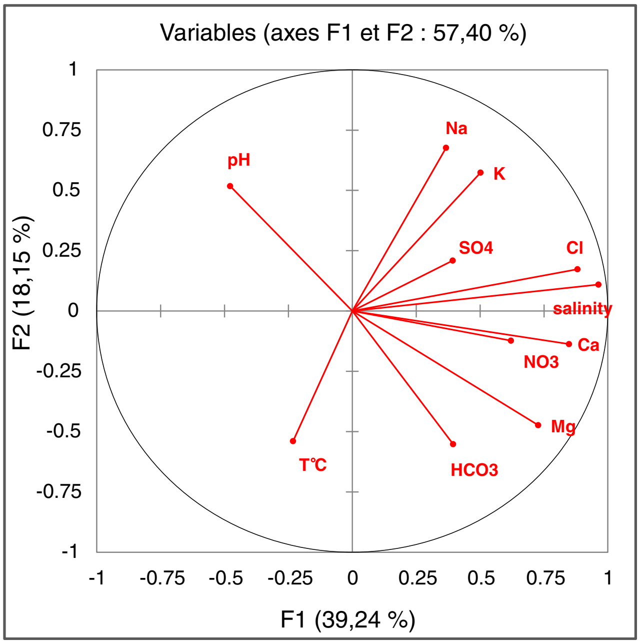


Fig. 10 Variable space deduced from the geochemical PCA of water resources in Grombalia basin

Statistical study: principal component analysis

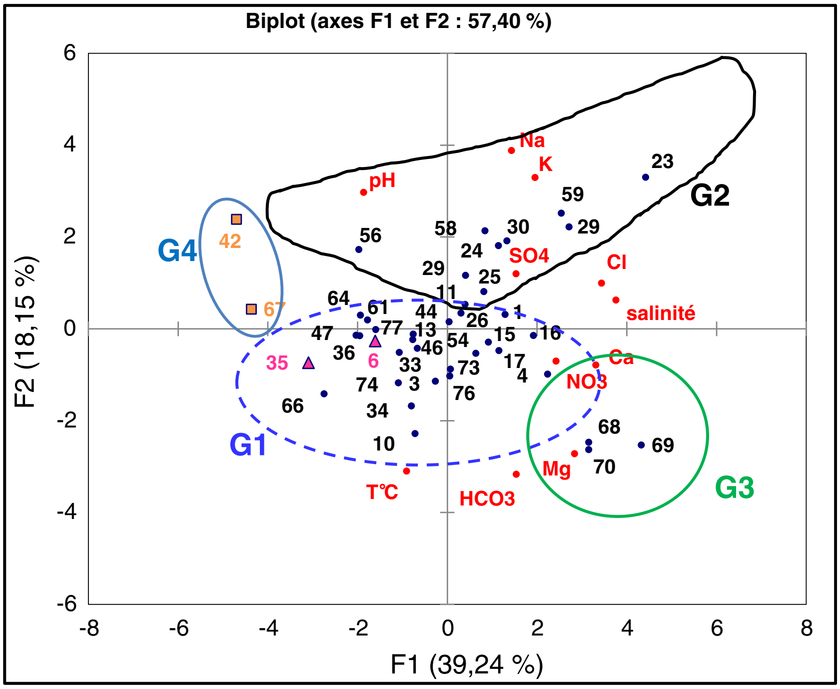
Principal component analysis (PCA) is the most common multivariate statistical method used in geochemistry studies (Lachaal et al. 2010). It used to analyzing relationships between the observed variables (Narany et al. 2014). In this study, PCA of chemical data was used to distinguish the contribution of natural and anthropogenic processes to the chemical composition of groundwater in Grombalia basin. The variables used for PCA were temperature, pH, salinity, Na^+, Ca^{2+}, K^+, Mg^{2+}, Cl^-, HCO_3^-, SO_4^{2-}, and NO_3^-.

Correlation matrix

The correlation matrix established between 11 hydrochemical variables, was used to determine the relationship existing between these variables (Table 3). This matrix indicates:

- A high correlation between Cl^-, Na^+, Mg^{2+}, and TDS, reflecting the significant contribution of these elements in mineralization of water;
- A low correlation between Na^+ and Cl^- ions ($r = 0.434$) shows that halite dissolution is affected by ion exchange process with the aquifer clay minerals;
- The high correlation between Ca^{2+} and Cl^- ($r = 0.761$), and Mg^{2+} and Cl^- ($r = 0.618$) indicates that both natural process (dissolution of minerals and ion exchange) contribute in the Grombalia groundwater mineralization;
- The positive correlation between Ca^{2+} and NO_3^- ($r = 0.644$) shows that groundwater nitrate contamination is caused by $Ca(NO_3)_2$ fertilizers.
- The low correlation between Ca and SO_4 shows that the dissolution of sulfate minerals is affected by the process of base exchange with clay minerals;
- A positive relationship between nitrate and TDS indicates the presence of SGW contamination related to the agriculture activities.

Fig. 11 Cluster analysis main sample groups according to their scores for F1 and F2 of water resources in Grombalia basin



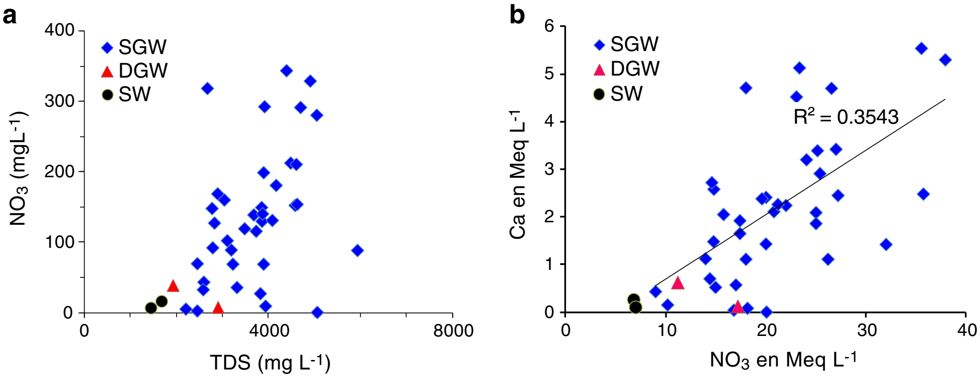


Fig. 12 NO_3/TDS and Ca/NO_3 relationships of water resources in Grombalia basin

Factors

PCA approach generates four main factors, which explained 57.40% of total samples variance: 39.24, 18.15, 10.24, and 9.82% for F1, F2, F3, and F4, respectively.

The salinity, Cl^-, Ca^{2+}, Mg^{2+}, and NO_3^-, showed a high positive loading for F1 (Table 4), whereas Na^+, K^+, pH, and temperature showed moderate positive and negative loading for F2, while HCO_3^- and SO_4^{2-} showed a moderate positive loading for F3 and F4, respectively.

F1 group consisting of salinity, Cl^-, Ca^{2+}, Mg^{2+}, and NO_3^- is largely influenced by natural and anthropogenic processes. This factor, with higher positive loading for NO_3^- and Ca^{2+} shows the existence of various anthropogenic pollutants, as $Ca(NO_3)_2$ fertilizers which is extremely used in agriculture regions. However, the high positive loading of Ca^{2+} and Mg^{2+} in F1 would be attributed to ion exchange reactions.

The second factor (F2), with moderate loading of Na^+ and K^+, indicates that the ion exchange process control the groundwater mineralization. In addition, this factor shows moderate loading for pH and temperature, indicating that these parameters are controlled by the natural process (impact of the atmospheric temperature) and anthropogenic activities (domestic and industrial wastewater).

The third factor (F3) is associated with a moderate loading for HCO_3^-. It indicates the low contribution of this element in the mineralization of the Grombalia groundwater.

Space variables

According the space variables diagram (Fig. 10), F1 takes high positive loading for all major elements except for Na^+ and K^+. This major factor describes mineralization of groundwater by both natural process (water/soil/rock interaction and ion exchange mechanism) and anthropogenic process (use of nitrogen fertilizer in agriculture).

F2 axis shows that the ion exchange process controlled the participation of Na^+ and K^+ elements in mineralization (Fig. 10). The negative loading for pH that corresponds to high positive loading for NO_3^- may provide insight into the significance of denitrification process in relation to the forest vegetation, relatively abundant in these regions (Ben Moussa et al. 2012).

Space samples groups

According to space sample diagram (Fig. 11), four groups of samples have been identified:

- The first group characterizes SGW and DGW samples with moderate mineralization. This group includes wells situated in the artificial recharge zone and in the south parts of Grombalia basin. This regions are characterized by moderately mineralized water and high temperatures (20 °C < T < 26 °C).
- The second group is placed on the positive side of F2 axis and groups the most mineralized samples taken in the North of region near Sebkat el Malah. This group is characterized by water with high pH and is slightly loaded with Na^+, Cl^-, K^+, and SO_4^{2-}, which explain the chloride-sodium facies in this region.
- The third group is situated on the positive side of F1 axis, is strongly associated with nitrate showing a high pollution related to agricultural practices. This group characterizes wells situated between Nianou and Turki towns. Indeed, the application of flood irrigation with over fertilization of cultivated perimeters led to the increase of nitrate, leaching through the return of irrigation water.
- The further group is placed on the negative side of F1 and the positive side of F2. This group characterizes surface water of Mejerda-Cap Bon channel, and indicates the good quality of this water type. It is strongly associated with pH and negatively correlated with nitrate, reflecting the importance of the phenomenon of denitrification. This phenomenon contributes to transformation of NO_3^- to N_2 by bacterial.

Groundwater nitrate contamination

SGW is characterized by high nitrate concentrations level, varying in a wide range, between 0.22 and 343.45 mg L^{-1}. The nitrate concentrations of most samples are above the drinking water standard (50 mg L^{-1}) (WHO 2006). The wells located in the central area are the higher contaminated wells by nitrate ($NO_3^- > 300$ mg L^{-1}), especially between Nianou and Turki towns. The high nitrate concentrations are caused by the return flow from irrigation. In addition, NO_3^- is correlates positively with TDS values indicating the significant contribution of nitrate in the mineralization process (Fig. 12). Furthermore, the positive relation between NO_3^- and Ca^{2+} ($r^2 = 0.354$) suggests the high use of $Ca(NO_3)_2$ fertilizers in the downstream and central of the basin (Fig. 12).

Conclusion

In this study, multi-tracer approach has been carried out to understand the hydrogeochemical functioning and to identify the different processes controlling the groundwater salinization of the coastal aquifer system of Grombalia using piezometric, ACP, and chemical analysis. As consequence of the lack of the regulation of the water use, a groundwater flow perturbation was recorded in the central part of the aquifer, where a piezometric rise was registered. The piezometric rise is caused by the excess irrigation return-flow.

In addition, an increase in water salinity and nitrate pollution was observed in this area. SGW samples were classified into Na-Cl, Ca-Mg-Cl, and Mg-Cl water types. SGW mineralization is caused by two origins: natural and anthropogenic processes. The natural mechanisms consist in the dissolution of halite, gypsum, and anhydrite, and the ion exchange. The anthropogenic processes are related to the intense agricultural activities mostly in the downstream and central parts of the Grombalia basin. The high nitrate contamination is caused by the combination of long-term flood irrigation practices and over use of fertilization.

Consequently, it seems necessary to implement a sustainable water management program in the region especially in the central part of the aquifer, which is based in the balance of multiple water sources use (SGW, DGW, dams, and water from the North-West).

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