

1 **Assessing seasonal variations and aquifer vulnerability in coastal aquifers of semi-arid**  
2 **regions using a multi-tracer isotopic approach: The case of Grombalia (Tunisia)**

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11 **Abstract**

12 The Grombalia aquifer (NE Tunisia) is an example of an important source of water supply for regional and  
13 national development, where the weak controls over abstraction, fertilizers applications and waste disposal,  
14 coupled with limited knowledge of aquifer dynamics, is triggering aquifer over-exploitation and water quality  
15 degradation. Assessing the key role of groundwater to water resources security is therefore of paramount  
16 importance to support new actions targeted to preserve water quality and quantity in the long-run. This study  
17 hence presents one of the first investigations targeted to a complete assessment of aquifer dynamics in the  
18 Grombalia aquifer. A multi-tracer hydrogeochemical and isotopic ( $\delta^2\text{H}$ ,  $\delta^{18}\text{O}$  and  $^3\text{H}$ ) approach permitted to  
19 study the influence of seasonal variations on piezometric levels, chemical and isotopic compositions and  
20 groundwater recharge. A total of 116 samples were collected from private wells and boreholes during a one year  
21 monitoring campaign (February-March 2014, September 2014 and February 2015). Results revealed the overall  
22 unsuitability of groundwater for drinking and irrigation purposes ( $\text{NO}_3 > 50 \text{ mg/L}$  in 51% of the wells;  $\text{EC} >$   
23  $1000 \mu\text{S/cm}$  in 99% of the wells). Isotopic balance coupled to piezometric investigation indicated the  
24 contribution of shallow aquifer to deep groundwater recharge. The study also revealed the weakness of business  
25 as usual management practices, highlighting possible solutions to tackle water related challenges in the  
26 Grombalia region where climate change coupled to population growth and intensive agricultural activities have  
27 generated a large gap between demand and available water reserves, hence becoming a possible driver for social  
28 insecurity.

29           **Keywords:** Quality assessment· Isotopes· Groundwater recharge· Mixing process· Tunisia

30   **1.       Introduction**

31   Almost one-third of the total area of the world is arid land (FAO 1989). This means that, around 20% of the total  
32   population lives in regions where water scarcity is often associated to famine and severe poverty (UN 2017),  
33   highlighting how aridity can have impacts that spans out of the environmental domain to the social one. In the  
34   long-term, and especially if climate change will persist to increase droughts in these regions, aridity will more  
35   and more become the driver of environmental migrations and of the related widespread of climate refugees. In  
36   fact, when aridity endures, freshwater inevitably becomes a limiting factor for crops, livestock and domestic  
37   consumption, while food security, in some cases already representing a serious concern, will become even more  
38   challenging (Robins and Fergusson 2014). Although, populations are trying to adapt to this situation, for many  
39   this will imply a conscious move to another place to survive. Such moves may spark conflict with other  
40   communities, as an increasing number of people will compete for a decreasing amount of resources (UNHCR  
41   2017). In this context, environmental migrants are defined as people who, “for reasons of sudden or progressive  
42   changes in the environment that adversely affect their lives or living conditions, are obliged to leave their  
43   habitual homes, or choose to do so, either temporarily or permanently, and who move either within their territory  
44   or abroad ” (IOC 2017). For example, Morocco, Tunisia and Libya each lose more than 1,000 Km<sup>2</sup> of productive  
45   land every year due to desertification (Prakash 2016) and residents on the edge of the Sahara Desert are forced to  
46   move to cities in the Maghreb (North Africa) and/or in Europe. Therefore, under these conditions it is  
47   fundamental to assess the role of renewable groundwater as the key to water resource security in semi-arid areas,  
48   and consequently to perform sound multidisciplinary investigations that can support new science-based  
49   management practices able to achieve its protection in the long-term. In fact, it is well known that in arid and  
50   semi-arid zones, like in the Middle East and North Africa (MENA region), groundwater often constitutes the  
51   main source of perennial water available for drinking, agricultural and industrial purposes (Zalidis et al. 2002;  
52   Re and Zuppi 2011; Bahir et al. 2012; Ouhamdouch et al. 2016), although the lack of adequate control over  
53   abstraction, fertilizers applications and waste disposal can lead respectively to aquifer over-exploitation (if  
54   withdrawal exceeds natural replenishment rates; Custodio 2002) and water quality degradation (Tropp and  
55   Jägerskog 2006; FAO 2011). Indeed, numerous studies revealed that major hydrological problems of arid or  
56   semi-arid rural zones in the MENA region are specifically associated to salinization, pollution and climate  
57   change. For example, the high salinity registered in the Souss basin (Southwest of Morocco) is explained by  
58   seawater intrusion and agricultural return flow, coupled with the decline of groundwater level. Together, these

59 processes pose serious problems to the agricultural and domestic water supplies and hamper the future  
60 groundwater availability (Bouchaou et al. 2008). In the Mediterranean side of Morocco, the Bou-Areg coastal  
61 aquifer is characterized by a high salinization, due to both natural processes (namely water-rock interactions) and  
62 human impacts due to agricultural return flows, making water unsuitable for irrigation practices and implying  
63 that farmers have to resort to alternative water resources, often with higher costs (Re et al. 2013; Re and Sacchi,  
64 2017). Similarly, the high groundwater mineralization of Chougafiya basin, situated in the central part of  
65 Tunisia, is controlled by water–rock interactions followed by ion exchange reactions and the mixing process  
66 with *Sebkha* (salt deposits), which consequent pressure on groundwater quality by becoming unsuitable for basic  
67 needs (Farid et al. 2013). In addition to natural and anthropogenic salinization and groundwater over-  
68 exploitation, climate change may exacerbate already existing water scarcity issues. For example, the El Daba’a  
69 shallow aquifer, located in north-west of Egypt, is currently facing a severe water crisis as a result of reported  
70 droughts affecting the Southern Mediterranean coast (Yousif et al. 2015). Consequently, as in many other  
71 countries of the MENA region, Tunisian authorities are becoming aware that water resources used to supply  
72 domestic and agricultural needs are limited (Gaaloul et al. 2014; Ayadi et al. 2016), and actions should be  
73 undertaken to prevent water crisis. The Grombalia coastal region, subject of the present study, is one of the most  
74 important agricultural areas in north-east Tunisia, which suffers from groundwater quality degradation and  
75 intensive exploitation. In fact, in this region, like in several developing countries, the water supply network is not  
76 available for all the households and hence most of the local inhabitants, especially in the rural areas, depend on  
77 groundwater for their basic needs. However, the often unregulated abstraction and widespread contamination  
78 often limits its suitability for drinking and irrigation purposes (Lachaal et al. 2016; Kammoun et al. 2018). This  
79 critical situation of water stress is the driver of several socio-economic issues, as discussed by Tringali et al.  
80 (2017): (i) decrease of crop production; (ii) farmers’ migration from rural areas towards cities in search of new  
81 activities, and (iii) poverty increase due to loss of agricultural work. Therefore, a good knowledge of Grombalia  
82 aquifer system is the main challenge for the development of such an important agricultural district and can be the  
83 driver for a new socio-economic development that takes into account both the environmental issues and needs of  
84 local populations (Re 2015). In particular, to assure a suitable groundwater management in the long-run, besides  
85 assessing the drivers of groundwater contamination, it is fundamental to reach a better knowledge of the aquifer  
86 system as a whole, also taking into account its responses to seasonal variations. In fact, despite several studies  
87 have already been carried out within this basin, the majority were only targeting the shallow aquifer, which is the  
88 most exploited and most contaminated (e.g. Hamza et al. 2010; Ben Moussa and Zouari 2011; Charfi et al.

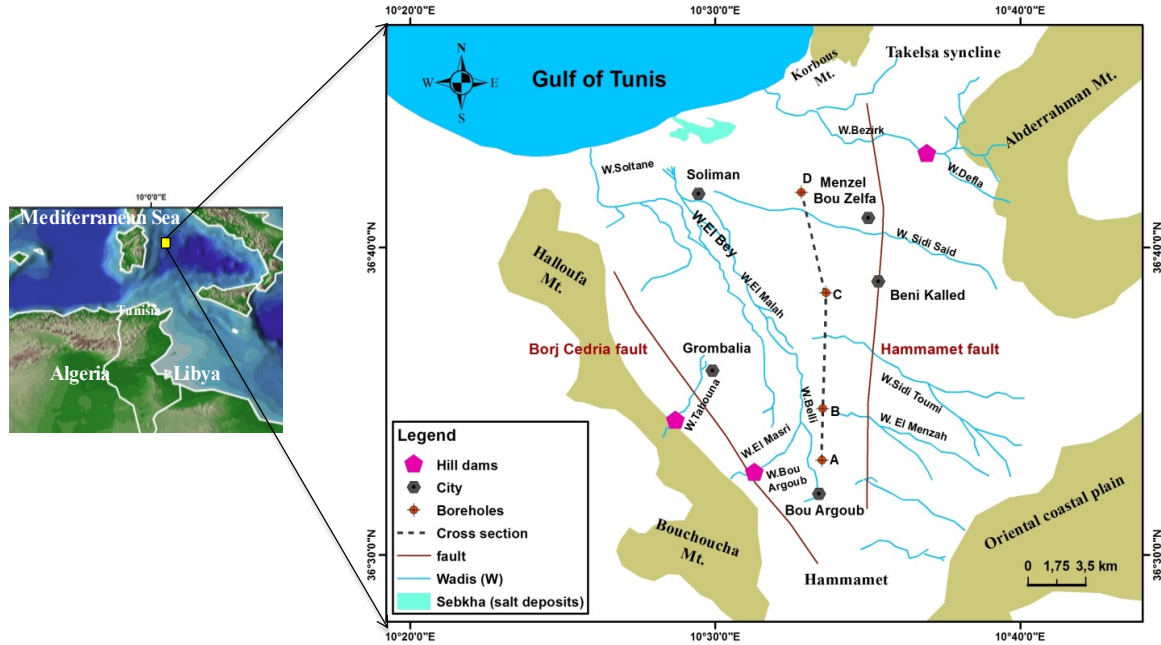
89 2013a; Tlili-Zrelli et al. 2013; Gaaloul et al. 2014). As a consequence, there is currently little knowledge on the  
90 deep aquifer and on the main processes regulating its chemistry and quality (Charfi et al. 2013b; Lachaal et al.  
91 2016; Re et al. 2017). Indeed, a complete assessment would permit to gain further understanding on the overall  
92 groundwater availability and its potential to sustain human activities and natural ecosystems.

93 This work therefore aims at (i) performing a complete groundwater quality assessment of the multilayer aquifer  
94 system of Grombalia using multi-tracer hydrogeochemical approach, (ii) assess the effect of seasonal variations  
95 on groundwater quality, and (iii) present a preliminary evaluation of the interactions between the shallow and  
96 deep aquifers. Overall this investigation can demonstrate the fundamental role of multidisciplinary  
97 hydrogeological assessment to support sustainable development in arid and semi-arid regions, highlighting the  
98 crucial role that adequate groundwater management can play to reduce poverty and environmental migrations.

## 99 **2. Site description**

100 Grombalia Basin is a part of the Cap Bon peninsula located in the north-eastern part of Tunisia (Fig. 1). It  
101 extends between latitudes 36°29'00"- 36°42'00"N and longitudes 10°27'00"- 10°47'00"E and covers a surface of  
102 719 Km<sup>2</sup>. It belongs to Northern hemisphere and according to specific weather conditions, the year is divided  
103 into four meteorological seasons: summer (June, July, August), fall (autumn: September, October, November),  
104 winter (December, January, February) and spring (March, April, May). The studied area is characterized by a  
105 semi-arid climate with an alternation of dry (summer and spring) and rainy (winter and fall) seasons, influenced  
106 by humid air masses coming from the Mediterranean Sea. The average annual precipitation in the region is  
107 approximately 506 mm/year, 60% of which occur from November to March, while it hardly rains in summer and  
108 early autumn seasons (CRDA; calculated over a period of 1954-2015). As concerns the period object of the  
109 present investigation, the mean precipitation recorded between 2014 and 2015 was about 458 mm (with monthly  
110 precipitation of 61 mm in February-March 2014, 13 in September 2014 and 106 mm in February 2015). The  
111 atmospheric temperature (T) is very low in winter (T = 7 °C) and very high in summer (T = 32 °C; CRDA 2015).  
112 The mean potential evapotranspiration is about 920 mm/year, varying in time and space depending on climatic  
113 parameters and surface occupation (i.e. vegetation cover, lake, river, bare soil, buildings; Bahir et al. 2012),  
114 therefore higher potential evapotranspiration is generally registered in July and August (CRDA 2015).

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**Fig.1** Localisation of the study region: Grombalia basin (North-east of Tunisia)

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From a geological point of view, Grombalia plain is defined as a graben characterized by NW-SE direction (Castany 1948). The graben structure was related to two normal faults, Borj Cedria (NNW–SSE) and Hammamet (NE–SW), that appeared during the Middle Miocene (Hadj Sassi et al. 2006).

121

The groundwater system of Grombalia is composed by shallow and deep aquifers which are separated by a thick clay layer (Ben Moussa and Zouari 2011). The shallow aquifer is filled by Quaternary sediments (alluvium, sands, and sandy clays) reaching to 50 m depth, whereas the deep aquifer is composed by Miocene series (Begli Formation), i.e. essentially by sandstone deposits (Fig. 2). These two aquifers are often communicating via sandy clay semi-permeable layers (Ennabli 1980; Hadj Sassi et al. 2006; Charfi et al. 2013b; Lachaal et al. 2016). Hydraulic conductivity values range from  $5.4 \times 10^{-6}$  (north and west of the plain) to  $6.5 \times 10^{-3}$  m/s (in the south and east part of the plain) according to Charfi et al. (2013). The aquifer transmissivity varies between  $25 \times 10^{-4}$  and  $2 \times 10^{-2}$  m<sup>2</sup>/s and the storage coefficient is equal to  $5.5 \times 10^{-3}$  (Tilil-Zrelli et al. 2013).

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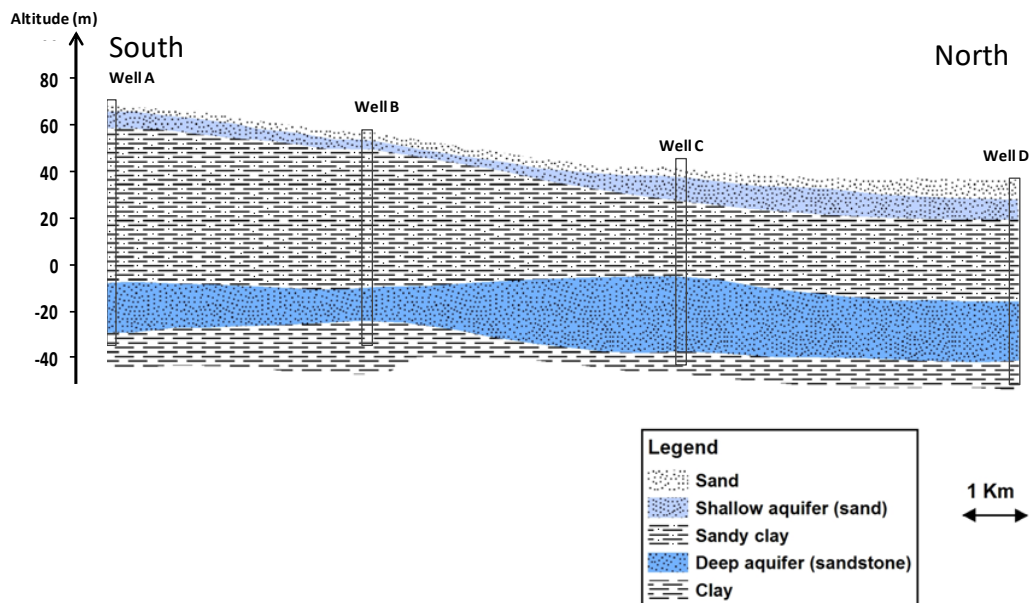
The hydrographical network in this region is well developed and composed of several rivers (*wadis*) with permanent or ephemeral regime. The most important river is Wadi El Bey, which constitutes the main water supply in this region. Given the relatively large amount of water provided by Wadi El Bey, two dams (Masri and Tahouna) were built to ensure a better management of surface water resources.

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In terms of economic activities, the studied region is primarily known for its intense agricultural activities. The latter include viticulture, arboriculture, cereals and livestock. Secondly, it is also characterized by its industrial

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135 activities, such as agro-food industries, textiles and dairy products. All activities are highly water-dependent, and  
 136 can possible cause water deficit in the region. Therefore, the Medjerda-Cap Bon canal was set up to ensure the  
 137 transfer of surface water from the north, i.e. from several dams built on the country's longest river, the Medjerda,  
 138 to Grombalia region. The transferred water has been used for irrigation and for artificial recharge in some wells  
 139 since 1992. Despite artificial recharge and irrigation by the Medjerda-canal water, the piezometric state of the  
 140 whole aquifer system is currently continuing to decline. This basin is being exploited by wells from 20 to 40 m  
 141 of depth. The exploitation of shallow aquifer was estimated at 106.0 Mm<sup>3</sup> in 2015, which exceeds (of almost the  
 142 double) renewable resources estimated at 51 Mm<sup>3</sup>/year (DGRE 2015). For the deep aquifer, the exploitation has  
 143 passed from 2.16 Mm<sup>3</sup> in 1990 to 21.62 Mm<sup>3</sup> in 2014, due to the increasing construction of deep boreholes (more  
 144 than 500 along the Grombalia plain) whose depths could reach to 160 m, while the renewable resources are  
 145 estimated to 9.5 Mm<sup>3</sup>/year (DGRE 2015). Therefore, both aquifers suffer from an over-exploitation of water  
 146 resources.  
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148

149 **Fig.2** Hydrogeological cross section (Data source: CRDA 2015)

150 **3. Materials and methods: data acquisition and methodology used**

151 Sampling of groundwater within the Grombalia region was carried out on a six-month basis during three  
 152 campaigns. A total of 116 samples were collected from the existing wells and boreholes (Fig. 3) at several

153 periods: (i) between February and March 2014 (spring 2014), (ii) in September 2014 (fall 2014) and (iii) in  
154 February 2015 (spring 2015). The second and third sampling campaigns were targeted to assess the occurrence  
155 of chemical and isotopic content evolution following seasonal variations. The spatial distribution maps of the  
156 sampling points were performed using the software package of Arc GIS (Ver. 9). Chemical analyses and isotopic  
157 measurements were conducted at the Radio-Analysis and Environment Laboratory of the National Engineering  
158 School of Sfax (Tunisia). Major elements ( $\text{Na}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{K}^+$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ ) were analysed by means of  
159 high performance ion liquid chromatography (HPLC). Carbonate and bicarbonate concentrations were analysed  
160 by titration using HCl acids (0.1N) and phenolphthalein and methyl red as colour indicators. The ionic balance  
161 for all samples was within  $\pm 5\%$ . Stable isotope ratio ( $^{18}\text{O}/^{16}\text{O}$  and  $^2\text{H}/^1\text{H}$ ) analyses were performed using the  
162 Laser Absorption Spectrometer LGR DLT 100 (Penna et al. 2010). Results are reported in ‰ vs. SMOW  
163 (Standard Mean Oceanic Water) with an analytical error  $\pm 1$  for  $\delta^2\text{H}$  and  $\pm 0.1$  for  $\delta^{18}\text{O}$ . Tritium analyses were  
164 performed using electrolytic enrichment and liquid scintillation counting (Taylor 1976). Tritium concentration is  
165 expressed in tritium unit (TU), according to isotope ratio  $^3\text{H}/^1\text{H} = 10^{-18}$ , with an analytical error equal to 0.3.

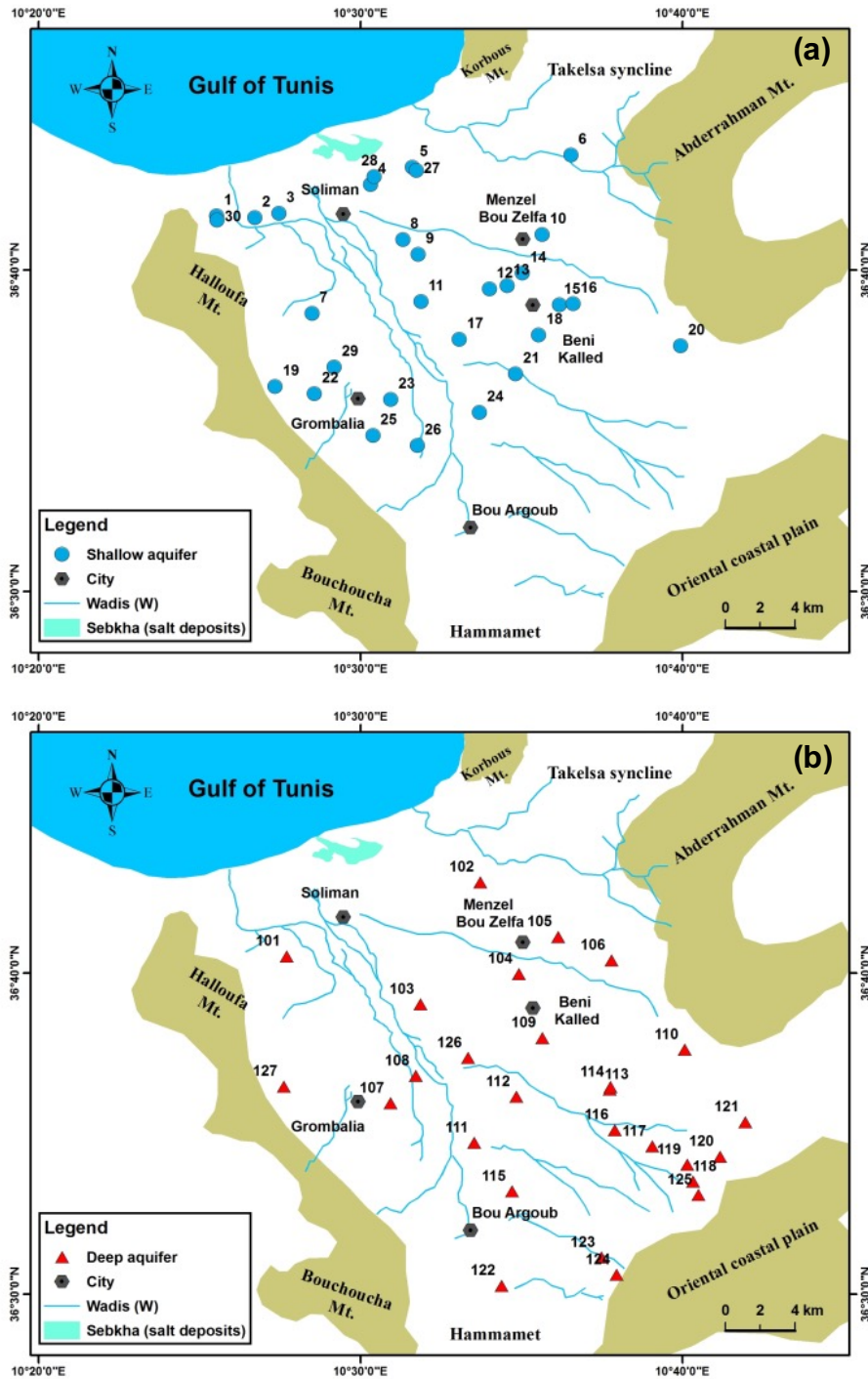


Fig. 3 Sampling map in the Grombalia basin for: **a** shallow aquifer; **b** deep aquifer

#### 4. Results and discussions

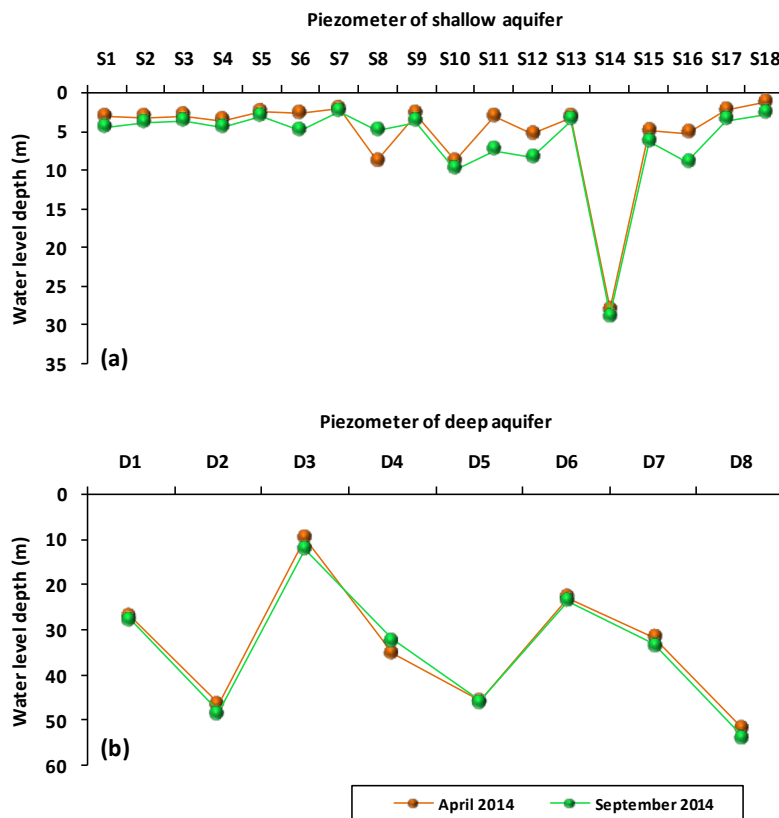
##### 4.1. Water table monitoring

Water table monitoring and piezometric studies were established based on available data for the year 2014 provided by the Regional Commissariat for Agricultural Development of Nabeul -Tunisia (CRDA 2015). The piezometric study aims to determine water level depth and to temporarily and spatially monitor its evolution.



173 Figure 4 shows that water level depth is season-dependent, increasing in rainy season (in April) and decreasing  
 174 in dry one (in September), for both aquifer layers, except for wells S8 and D4 from shallow and deep aquifers  
 175 respectively. These two samples, located close to artificial recharge sites, recorded a slight water level rise with  
 176 less than one meter. For the shallow groundwater, the average fluctuation is about 1.2 m with the maximum  
 177 decrease (4.3 m) observed in well S11 and the maximum increase reach to 3.8 m in well S8. Whereas for the  
 178 deep aquifer, the maximum decrease is about 2.9 m (well D4) and the maximum increase reach to 2.3 m (well  
 179 D3) with an average fluctuation of about 0.7 m. Therefore, seasonal fluctuations of water level depths are higher  
 180 in shallow aquifer rather than in the deep one. This could be related to the variability of precipitations, recharge  
 181 and exploitation rate.

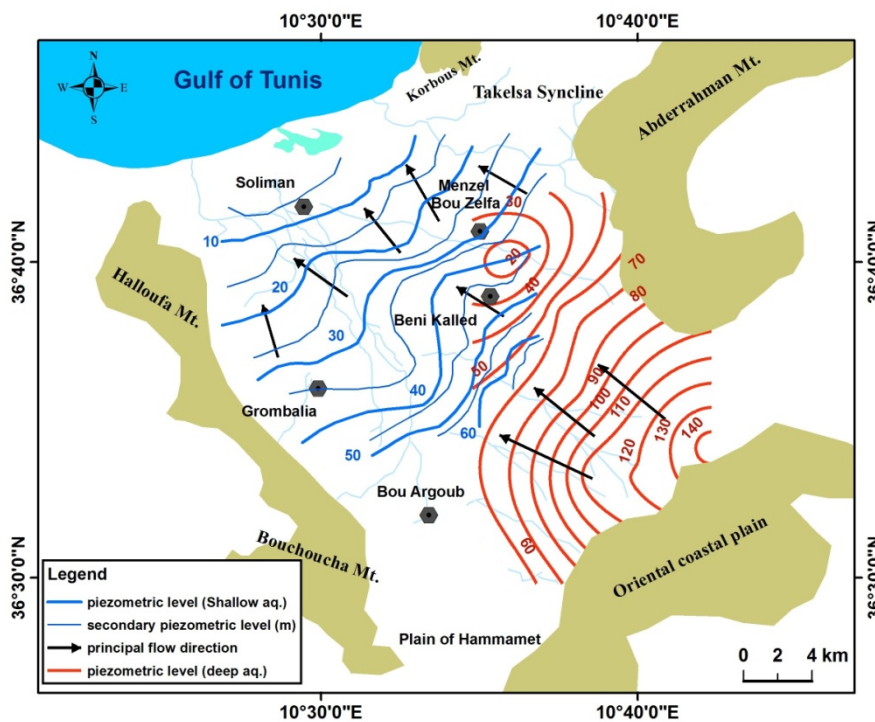
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184 **Fig. 4** Seasonal fluctuation of water levels depths in 2014 for: **a** shallow aquifer; **b** deep aquifer (CRDA 2015)

185 The study of the groundwater piezometry in Grombalia basin, for shallow and deep aquifers, permitted to  
 186 identify groundwater flow direction, recharge zones and the interconnection between these two aquifer layers.  
 187 For both aquifers, piezometric levels decrease from the south towards the north of the plain, passing from 60 to  
 188 10 m for the shallow aquifer and from 140 to 20 m for the deep one (Fig. 5). Two main recharge zones can be  
 189 identified: one is located in the east of the plain, corresponding to foothills of Abderrahman Mountain, and one  
 190 in the south-east part, close to the Bou Argoub region, resulting in a main SE-NW groundwater flow direction,  
 191 hence from upstream towards downstream of the basin (the depression of Sebkhah El Malah and the Gulf of  
 192 Tunis). In Beni Kalled and Menzel Bou Zelfa regions, the piezometric curves are tighter, reflecting a strong  
 193 hydraulic gradient which can be explained by high groundwater abstraction rates in these areas. Furthermore,  
 194 downstream of the basin, the piezometric curves become more spaced with a low hydraulic gradient due to the  
 195 decrease in exploitation of the deep aquifer. In addition, the piezometric map (Fig. 5) highlights the possible  
 196 recharge of the deep aquifer by shallow groundwater, driven by the lithology and the structure of the  
 197 intermediate formations, and/or in presence of existing micro faults contributing to this direct recharge.



198  
 199 **Fig. 5** Piezometric map of shallow aquifer (*blue lines*) and deep aquifer (*red lines*) in 2014 (Data source: CRDA  
 200 2015)

201 **4.2. Mineralization origin and quality assessment**

202 The physico-chemical data of the analysed groundwater samples (Table 1 and 1S in Electronic supplementary  
 203 material) show a relatively wide range of temperature values. For the first campaign of spring 2014, temperature

204 values range from 15.7 to 21.3 °C (mean 18.2 °C) in the shallow aquifer and from 15.8 to 23.7 °C (mean 20.4  
205 °C) in the deep aquifer. For the second and third sampling campaigns, temperature values decreased slightly in  
206 spring and increased in fall season. This variation is related to the water level depths and the influence of  
207 atmospheric temperature. The pH values are generally neutral for all samples collected from both aquifers during  
208 all campaigns. The average electrical conductivity (EC) for the shallow groundwater samples, collected in spring  
209 2014, is about 3867.7  $\mu\text{S}/\text{cm}$  varying within a wide range of values, from 1039  $\mu\text{S}/\text{cm}$  (well 23, located in the  
210 south-western part of the plain) to 9180  $\mu\text{S}/\text{cm}$  (well 1, the nearest well to the Mediterranean Sea). For the deep  
211 aquifer, EC values range from 1042 (well 122) to 14020  $\mu\text{S}/\text{cm}$  (well 108), with an average value equal to  
212 2640.1  $\mu\text{S}/\text{cm}$ . These values confirm the high mineralization of groundwater in Grombalia basin. Variation of  
213 water quality is a function of physico-chemical parameters and chemical composition that are greatly influenced  
214 by both the geological nature of the reservoir and anthropogenic activities (Subramani et al. 2005).

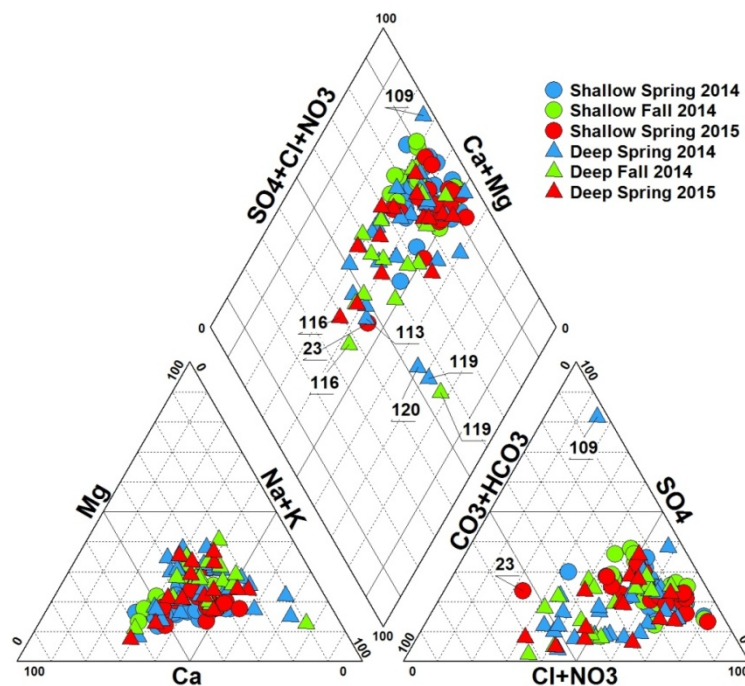
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217 **Table 1** Descriptive statistics of chemical and isotopic parameters. Concentrations of major ions are expressed in mg/L; water hardness is expressed as milligrams of calcium  
 218 carbonate equivalent per litre (mg/L of CaCO<sub>3</sub>); isotopic values as ‰ vs. SMOW and tritium values are expressed in TU.

		T	pH	EC	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	CO <sub>3</sub> <sup>2-</sup>	HCO <sub>3</sub> <sup>-</sup>	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	TDS	Hardness	δ <sup>2</sup> H	δ <sup>18</sup> O	Tritium
		°C		µS/cm	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	‰ vs. SMOW	‰ vs. SMOW	TU
Shallow Spring 2014	Min.	15.7	6.9	1039.0	112.4	0.0	145.3	0.0	164.7	111.1	0.0	26.1	69.0	715.4	280.0	-37.1	-5.4	0.6
	Max.	21.3	7.8	9180.0	2932.4	514.7	759.6	0.0	481.9	1193.7	40.3	292.3	775.2	6357.5	3140.0	-20.8	-3.5	5.6
	Mean	18.2	7.2	3867.7	838.8	148.0	434.1	0.0	332.5	397.5	16.8	99.9	349.5	2617.0	1283.0	-28.4	-4.6	2.4
	Std. dev	1.4	0.2	1538.1	534.1	140.9	165.0	0.0	75.8	220.0	10.7	54.2	156.8	1110.9	569.0	3.2	0.5	1.2
Shallow Fall 2014	Min.	20.0	6.7	1872.0	290.0	2.2	216.5	0.0	195.2	143.4	3.8	46.5	151.6	1197.9	570.0	-32.1	-5.2	1.3
	Max.	25.8	7.4	7400.0	2101.9	444.8	847.7	12.0	486.7	864.9	37.4	221.2	610.3	4556.8	2150.0	-17.5	-2.1	2
	Mean	22.8	6.9	3871.4	796.3	137.5	464.0	0.7	319.8	373.7	16.4	111.5	338.2	2558.0	1303.0	-27.9	-4.5	2.8
	Std. dev	1.7	0.2	1517.5	464.0	141.9	193.0	3.0	86.8	203.1	9.5	53.7	138.7	963.7	491.0	3.5	0.7	0.7
Shallow Spring 2015	Min.	13	6.57	714	57.59	0.0	82.125	0.0	164.7	68.15	4.9	17.185	50.035	524.925	200.0	-35.3	-5.8	-
	Max.	21.2	7.7	9560.0	2889.3	430.5	695.5	0.0	445.3	1201.9	40.5	257.1	621.7	5967.5	2610.0	-25.0	-3.7	-
	Mean	17.0	7.0	3818.6	876.8	114.7	423.5	0.0	312.4	425.8	14.4	98.9	304.0	2570.4	1166.0	-28.7	-4.8	-
	Std. dev	2.3	0.3	2013.5	623.4	141.8	174.1	0.0	74.3	264.0	9.3	57.5	152.7	1268.4	571.0	2.4	0.5	-
Deep Spring 2014	Min.	15.8	7.0	1042.0	107.3	2.0	23.8	0.0	128.1	72.8	4.2	12.7	31.6	727.2	230.0	-34.8	-6.1	0
	Max.	23.7	7.7	14020.0	3436.0	230.6	3105.0	42.0	688.0	1888.4	63.6	557.3	696.5	10177.4	4030.0	-28.7	-4.5	2.4
	Mean	20.4	7.2	2640.1	473.3	55.7	383.0	4.8	321.3	275.9	16.1	89.7	181.8	1801.5	823.0	-31.8	-5.4	1.0
	Std. dev	1.4	0.2	2587.8	650.6	62.1	777.1	11.1	99.9	359.0	15.0	114.0	157.6	1938.0	840.0	1.7	0.4	1.0
Deep Fall 2014	Min.	22.0	6.7	1089.0	106.0	1.0	9.8	0.0	244.0	73.8	3.5	15.8	34.7	628.8	200.0	-34.3	-6.2	1
	Max.	27.7	7.4	5310.0	1214.8	148.1	640.1	36.0	396.5	511.3	22.4	160.9	269.7	3145.5	1330.0	-28.2	-4.6	1.1
	Mean	23.9	7.0	2156.3	358.5	30.6	201.1	11.3	316.9	202.6	11.6	68.0	134.5	1335.0	615.0	-31.0	-5.5	1.2
	Std. dev	1.5	0.2	1100.3	289.4	38.4	174.0	14.7	46.3	118.7	6.0	40.4	78.2	676.2	344.0	1.4	0.4	0.1
Deep Spring 2015	Min.	11.4	6.9	1077.0	112.5	0.0	30.1	0.0	140.3	83.7	3.1	11.4	65.2	755.8	350.0	-33.4	-5.9	-
	Max.	21.1	7.5	5120.0	1230.7	217.8	580.2	0.0	457.5	647.3	23.4	161.7	345.7	3263.0	1330.0	-28.5	-4.5	-
	Mean	17.1	7.1	2299.2	424.9	46.2	200.7	0.0	342.0	212.4	9.4	66.0	161.8	1463.4	677.0	-31.2	-5.4	-
	Std. dev	3.4	0.2	1045.9	285.9	59.1	155.8	0.0	83.1	143.2	5.1	35.6	82.2	651.6	292.0	1.6	0.4	-

220 The chemical compositions of analysed samples were plotted on Piper diagram in order to specify the different  
 221 groundwater types for both the shallow and the deep aquifers, and to assess their seasonal evolution (Fig. 6). The  
 222 majority of the samples collected in the shallow aquifer belong to the mixed pole in the triangle of cations,  
 223 whereas some groundwater samples present a slight dominance either of calcium or sodium. In the triangle of  
 224 anions, the same variability can be highlighted, with predominance either of chloride, bicarbonate or sulphate.  
 225 Therefore, based on the distribution of samples in the Piper diagram and the calculated percentage of each ion, it  
 226 emerges that for the shallow aquifer, there is an absence of dominant *facies*. For the shallow aquifer,  
 227 groundwater can be classified as mixed water type (76%), Ca(Na)-Cl (17%) and Na(Ca)-Cl (7%). On the other  
 228 hand, for the deep aquifer, the mixed water type constitutes 86% of the samples, although some samples display  
 229 either a Ca(Na)-Cl (5%) or Na(Mg)-Cl (4%) water type. Additionally, for some samples located in proximity of  
 230 the recharge zone (i.e. the foothills of Abderrahman Mountain), sodium and bicarbonate are the dominant ions  
 231 Na-HCO<sub>3</sub> (5%).  
 232 The difference of water type from one region to another could be explained by an evolution according to transit  
 233 time and/or the transition of formations' lithology, passing from continental deposits (in the south) to marine  
 234 deposits close to Gulf of Tunis (Charfi et al.2013b). The similarities of chemical *facies* for some samples  
 235 collected from shallow and deep aquifers suggest a possible groundwater mixing (Re et al. 2017). The Piper  
 236 diagram also highlights the absence of significant influence on groundwater types of seasonal variations. The  
 237 latter may affect ion concentration, but without changing the overall water type.

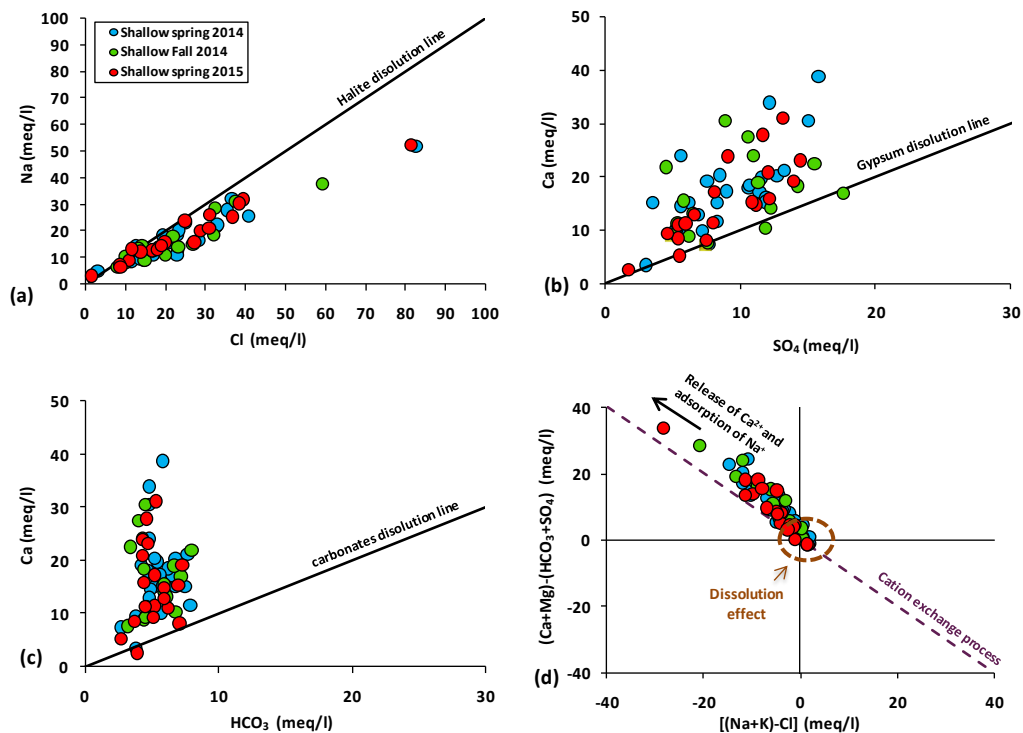


238  
239 **Fig. 6** Piper diagram of the Grombalia aquifer system

240 In fact, the evolution of groundwater chemical composition for some samples from both shallow and deep  
 241 aquifers, (Fig. 7 and Fig. 8) highlights the variability of major elements concentrations from one season to  
 242 another, that mainly depends on atmospheric temperature, controlling evaporation process, aquifer recharge and  
 243 return flow of irrigation water (Ben Moussa et al. 2011).

244 The principal process contributing to aquifer mineralization, as discussed by several authors (Ben Moussa and  
 245 Zouari 2011; Charfi et al. 2013; Lachaal et al. 2016; Re et al. 2017; Kammoun et al. 2018) and also confirmed by  
 246 the present survey, is the dissolution of evaporates (such as halite: NaCl, gypsum: CaSO<sub>4</sub>·2H<sub>2</sub>O and anhydrite:  
 247 CaSO<sub>4</sub>) generally existing in sedimentary deposits. These dissolution processes can be highlighted by both the  
 248 strong correlation between sodium *versus* chloride (Figs 7a, 8a) and calcium *versus* sulphate (Figs 7b, 8b), and  
 249 also by the under-saturation state for most of samples with respect to halite, gypsum and anhydrite. Figures 7c  
 250 and 8c show a poor correlation between calcium and bicarbonate indicating the inability of water to dissolve  
 251 carbonate minerals.

252 Thus, calcium excess and sodium deficiency characterizing some samples suggest that other mineralization  
 253 processes should be considered, such as the ion exchange reactions that significantly affect water chemical  
 254 composition, by which the Na<sup>+</sup> minerals are adsorbed on the surface of clay minerals against the release of Ca<sup>2+</sup>.  
 255 The ion exchange process was confirmed by the relation between [(Ca<sup>2+</sup>+ Mg<sup>2+</sup>) - (HCO<sub>3</sub><sup>-</sup> + SO<sub>4</sub><sup>2-</sup>)] and [Na<sup>+</sup> +  
 256 K<sup>+</sup> - Cl<sup>-</sup>], as reported by Garcia et al. (2001), (Figs 7d, 8d).



257

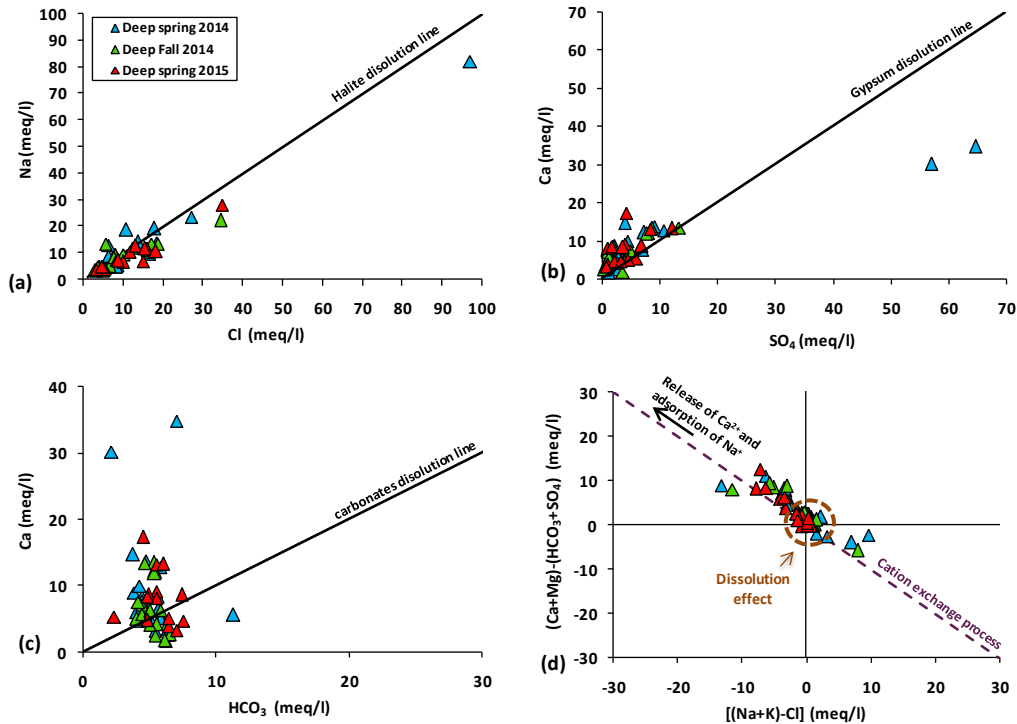
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259

**Fig. 7** Relationship between major elements for shallow aquifer samples: **a** Na versus Cl; **b** Ca

260

versus SO<sub>4</sub>; **c** Ca versus HCO<sub>3</sub>; **d** (Na + K) – Cl versus (Ca + Mg) - (SO<sub>4</sub> + HCO<sub>3</sub>)



261

**Fig. 8** Relationship between major elements for deep aquifer samples: **a** Na versus Cl; **b** Ca versus

262

SO<sub>4</sub>; **c** Ca versus HCO<sub>3</sub>; **d** (Na + K) – Cl versus (Ca + Mg) - (SO<sub>4</sub> + HCO<sub>3</sub>)

263

264

In addition, previous studies focusing on nitrate pollution (Hamza et al. 2010; Chenini et al. 2015; Re et al. 2017;

265

Ben Moussa and Zouari 2011; Charfi et al. 2013a) highlighted their significant influence on groundwater

266

salinization and quality degradation. As concerns the spring 2014 campaign, results shows that there are about

267

70% of the shallow aquifer's samples and 41% from the deep one that exceed the statutory limit for drinking (50

268

mg/L; WHO 2011). The highest nitrate concentrations are especially found in regions where agricultural

269

activities are dominant, hence associated to the high potential for contamination due to agricultural return flow

270

(Kammoun et al. 2018). On the other hand, relatively low nitrate concentrations are observed in the deeper wells

271

which can be explained by denitrification process that occurred in anaerobic condition. Several studies have

272

shown the agricultural source of contamination in this region (Chenini et al. 2015; Lachaal et al. 2016) whereas,

273

Zouari et al. (2015) and Re et al. (2017) revealed that besides agricultural activities (fertilizers and manure

274

application), nitrates are also generated by urban/domestic activities, due to the lack of adequate sanitation

275

systems in some parts of the plain.

276 In addition to nitrate pollution, high values of electrical conductivity make this water in doubtful suitability for  
 277 irrigation use. Analytical data for the three sampling campaigns show that 67% of the total samples collected  
 278 from shallow aquifer and 20% from the deep one are characterized by  $EC > 3000 \mu\text{S}/\text{cm}$ , therefore they are  
 279 considered as unsuitable for irrigation use. Also, the high values of calcium that characterise most of samples,  
 280 can affect water hardness and water suitability to both human consumption and irrigation. For example, the  
 281 calculated hardness values of samples collected in spring 2014 are very high, ranging from 280 (well 23) to 3140  
 282 mg/L (well 1) for shallow aquifer and from 230 (well 120) to 4030 mg/L (well 108) for the deep one. This could  
 283 be explained by water-rock interaction processes, as water flowing through aquifer formations' may dissolve  
 284 calcium and magnesium minerals. In addition, the excessive use of  $\text{Ca}(\text{NO}_3)_2$  and  $\text{MgSO}_4$  fertilizers may  
 285 increase water hardness, highlights the indirect role of nitrate fertilizers, inducing an acidic perturbation of the  
 286 solution which is buffered by carbonate dissolution, leading to an increase in water hardness (Spruill et al. 2002).  
 287 According to these high levels, the consumption of this water can have negative consequences for human health.  
 288 Since groundwater resources are mainly used for irrigation in the study area, monitoring of water suitability for  
 289 irrigation is crucial. Therefore, the different irrigation parameters: sodium percentage (%Na), permeability index  
 290 (PI), magnesium adsorption ratio (MAR), and sodium adsorption ratio (SAR) are calculated according to the  
 291 following equations (1, 2, 3 and 4):

$$292 \quad \%Na = \frac{(Na+K)}{(Ca+Mg+Na+K)} * 100 \quad (1)$$

$$293 \quad PI = \frac{(Na + \sqrt{HCO_3})}{Ca+Mg+Na} * 100 \quad (2)$$

$$294 \quad MAR = \frac{Mg}{Ca+Mg} * 100 \quad (3)$$

$$295 \quad SAR = \frac{Na}{\sqrt{\frac{Ca+Mg}{2}}} \quad (4)$$

296 Where, all ion concentrations are expressed in meq/L (Table 1S in Electronic supplementary material and Table  
 297 2).

298 Regarding sodium percentage values, all groundwater samples are permissible for irrigation except two samples  
 299 (wells 119, 120) with %Na falling between 60 and 80%. Concerning the permeability index, all water samples  
 300 with PI ranging between 0 and 75 are considered to be good and suitable for irrigation (Doneen 1964).  
 301 Analytical data show that just two groundwater samples from deep aquifer (wells 119, 120) were not suitable for  
 302 irrigation based on permeability index, whereas the rest of samples were good. In the study region, MAR values

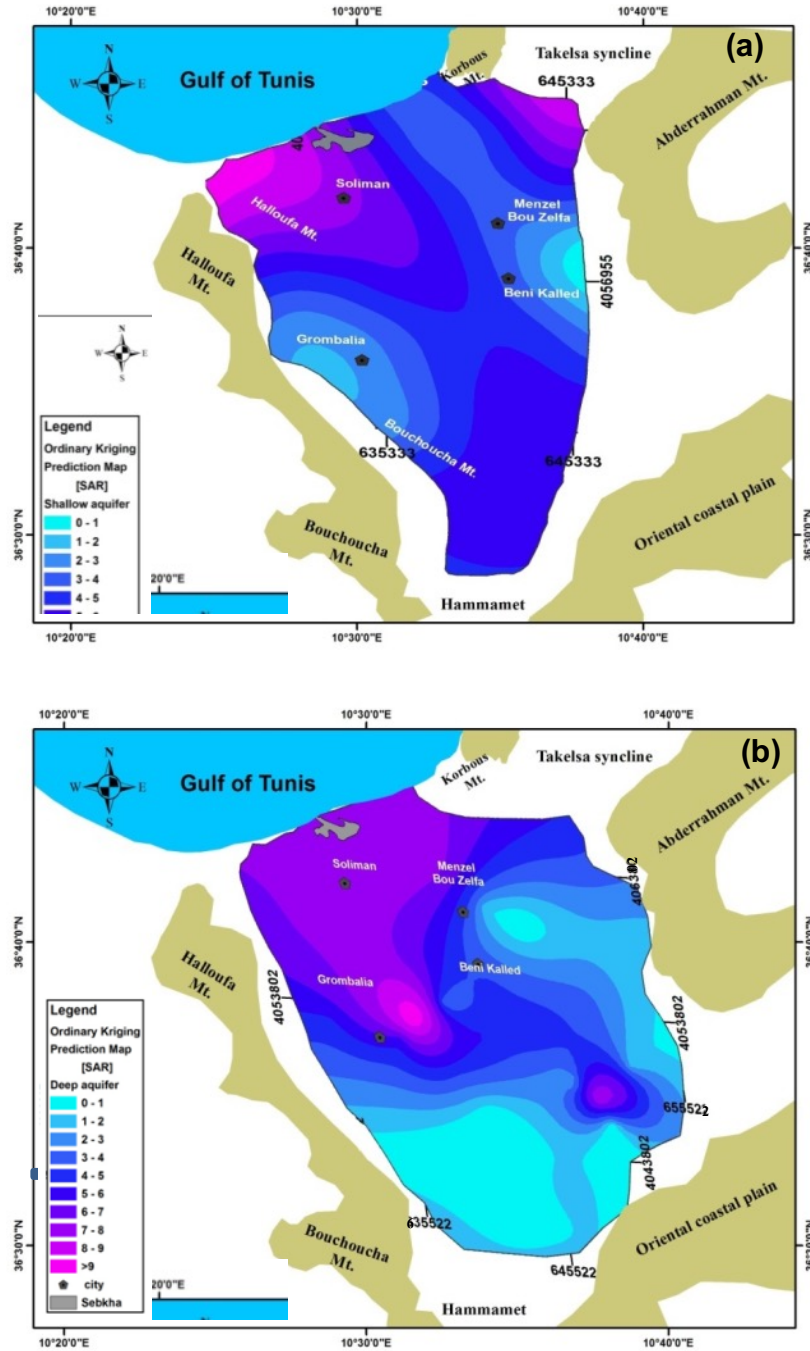


303 were found within a wide range. One groundwater sample (well 29) from shallow aquifer and ten samples from  
304 the deep one (wells 103, 108, 111, 113, 115, 116, 117, 119, 120, and 124), exceed the admissible limit of 50, that  
305 is considered harmful and unsuitable for irrigation purpose (Subramani 2005). It is important to evaluate soil  
306 exchange reactions and the probability of accumulation of sodium in the soil. For this purpose, Sodium  
307 Adsorption Ratio (SAR) is used to classify sodium hazards in irrigation water (Richards 1954).  
308 The calculated SAR values of groundwater samples from the shallow aquifer, collected in spring 2014, range  
309 from 2.8 to 9.3. Some groundwater samples are characterized by low SAR values (wells 10, 16, 19, 20 and 23)  
310 but after taking into account their high electrical conductivity, these samples are considered as unsuitable for  
311 irrigation, especially in soils with restricted drainage (Subramani et al. 2005) and harmful for plants and crops, as  
312 the perennial woody species in Grombalia (e.g. orange, lemon, grapes), that have low to moderate salt tolerance.  
313 The majority of samples show very high water salinity with average values of SAR. This type of water can be  
314 used, under favourable drainage conditions, just for irrigation of semi-tolerant and tolerant salt cultures. Only  
315 three samples represent waters with very high salinity and high SAR (wells 1, 3 and 8). These samples are  
316 considered to be poor water quality and therefore unsuitable for irrigation, particularly on clay soils. For the deep  
317 aquifer, SAR varies between 1.6 and 12.9 of which a single sample (well 108) is greater than ten. The spatial  
318 distribution of SAR parameter (Fig 9), produced by ordinary kriging technique, shows that the higher values  
319 characterized samples located in the north of the plain (Soliman region), towards the sea, for both aquifers. For  
320 the deep aquifer, high SAR values also occur in Grombalia and Beni Kalled regions.

321

322

323



324

325 **Fig. 9** Spatial distribution map of sodium adsorption ratio (SAR) produced by *ordinary kriging* method for: a  
 326 shallow aquifer (spring 2014); b deep aquifer (spring 2014)

327 **Table 2** Classification of irrigation water based on sodium adsorption ratio (SAR), magnesium adsorption ratio  
 328 (MAR), sodium percentage (%Na) and permeability index (PI) and their variation under seasons

		SAR	MAR	%Na	PI
Shallow Spring 2014	Min.	2.8	18.0	26.3	31.5
	Max.	9.3	44.1	53.0	65.1

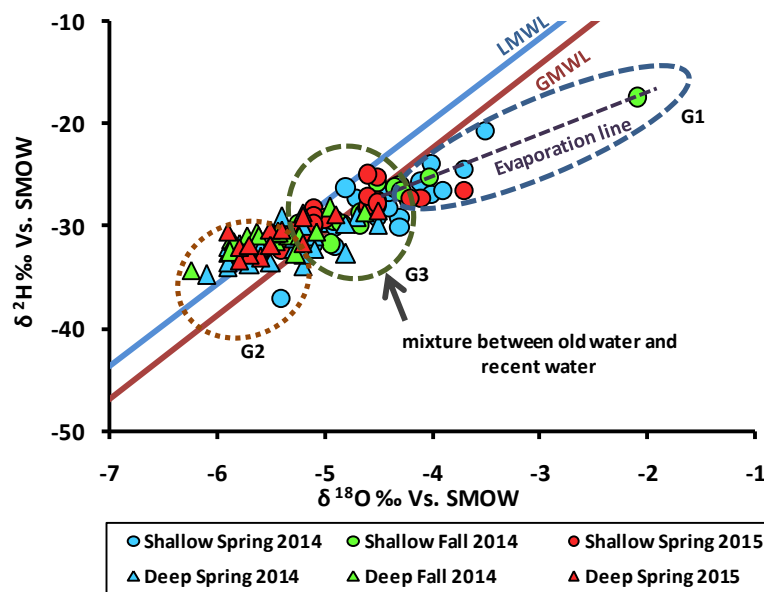
	Mean	4.7	32.2	40.6	46.3
	Min.	2.4	18.4	28.1	32.6
Shallow Fall 2014	Max.	8.1	49.0	49.6	56.2
	Mean	4.4	35.1	38.3	44.0
	Min.	2.1	18.8	33.3	37.7
Shallow Spring 2015	Max.	10.2	52.1	55.6	71.8
	Mean	5.2	34.9	43.9	50.8
	Min.	1.6	12.0	26.3	26.6
Deep Spring 2014	Max.	12.9	66.0	71.6	84.7
	Mean	3.9	43.3	41.1	52.4
	Min.	1.6	15.5	28.5	37.7
Deep Fall 2014	Max.	9.5	65.4	77.4	91.5
	Mean	3.6	46.0	42.2	54.7
	Min.	1.7	10.4	29.2	39.9
Deep Spring 2015	Max.	7.7	59.7	55.0	65.1
	Mean	3.4	40.9	39.7	51.4

329

### 330 4.3. Isotope composition of groundwater and recharge process

331 Stable isotopes of water molecule ( $\delta^2\text{H}, \delta^{18}\text{O}$ ) are indicators of local conditions present at the moment of  
332 groundwater recharge (Faure 1986), and used to evaluate the physical processes affecting water masses, such as  
333 mixing and evaporation processes (Geyh 2000). The isotopic compositions of groundwater samples collected  
334 from the shallow aquifer range from -5.4 to -3.5‰ vs. SMOW for  $\delta^{18}\text{O}$  and from -37.1 to -20.8‰ vs. SMOW for  
335  $\delta^2\text{H}$  (spring 2014). Isotopic signatures of the majority of groundwater sample are comparable to those of the  
336 local precipitation, -4.3 for  $\delta^{18}\text{O}$  and -24.8‰ vs. SMOW for  $\delta^2\text{H}$ , in Tunis-Carthage meteoric station  
337 (N°6071500) according to the global network of isotopes in precipitation (GNIP 2007). This similarity indicates  
338 the aquifer recharge through rapid infiltration of rainwater. In the deep aquifer, isotopic values range from -6.1 to  
339 -4.5‰ vs. SMOW for  $\delta^{18}\text{O}$  and from -34.3 to -28.2‰ vs. SMOW for  $\delta^2\text{H}$  (spring 2014). It is seen from Fig.10  
340 that most samples from both shallow and deep aquifers fall around the GWML (Global Water Meteoric Line)  
341 reported by Rozanski et al. (1993) according to the equation  $\delta^2\text{H} = 8.17 \delta^{18}\text{O} + 10.35$  and the Local Meteoric  
342 Water Line of Tunis-Carthage (LMWL) with  $\delta^2\text{H} = 8 \delta^{18}\text{O} + 12.4$  reported by Zouari et al. (1985). This indicates  
343 that they originated from modern precipitation infiltration. According to the position of the points on the  $^{18}\text{O}/^2\text{H}$   
344 diagram three groups of groundwater have been identified. The first group (G1 in Fig.10) is characterized by the  
345 most enriched samples collected from shallow aquifer highlighting the significance of surface water input in the  
346 evolution of the observed isotopic composition, and/or the return flow of irrigation water affected by evaporation  
347 process. These samples generally present relatively high tritium values (> 2 TU) and high nitrate contents (> 50  
348 mg/L). The second group (G2) represents the most depleted water samples, mainly from the deep aquifer,

349 characterized by a different isotopic composition than present day precipitation.. This could be explained by the  
 350 presence of old water recharged in a different period in the past. This hypothesis is supported by the very low  
 351 tritium values characterizing these samples (Table 3).  
 352 Finally, a third group (G3) can be representative of mixing process occurring between recent and old  
 353 groundwater. This explains the position of some points between the depleted and the relatively enriched stable  
 354 isotopes, respectively representing the old and present day recharge water. Stable isotopes of the water molecule  
 355 are related to the isotopic signature of the precipitation from which they derive, taking into account the  
 356 atmospheric circulation which causes these masses of air that dominate well-defined seasons. In fact, rain is  
 357 more depleted in winter and more enriched in summer. In addition, the fact that these two aquifer layers display  
 358 similar signatures of  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$ , especially for the third group, is a sign of water exchange between these two  
 359 aquifers especially in the eastern part of Grombalia region (wells 8, 16, 104, 105, 110, 113, 114, 122, 126).  
 360

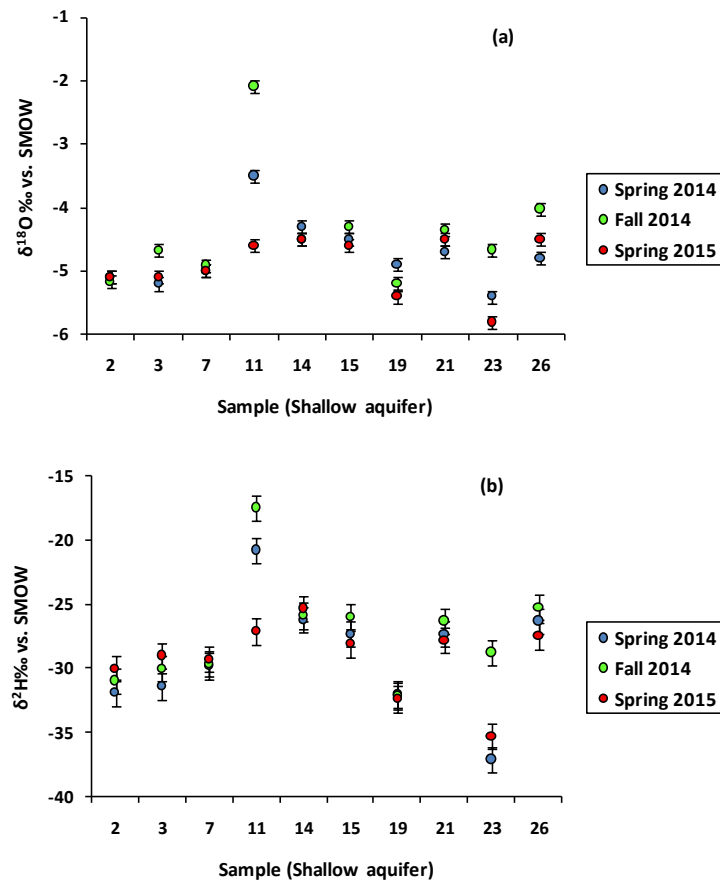


361  
 362 **Fig. 10** Plot of  $\delta^2\text{H}$  versus  $\delta^{18}\text{O}$  of the shallow and deep groundwater samples during three campaigns, with  
 363 *GMWL* corresponds to Global Water Meteoric Line reported by Rozanski et al. (1993) according to the equation  
 364  $\delta^2\text{H} = 8.17 \delta^{18}\text{O} + 10.35$  and *LMWL* corresponds to Local Meteoric Water Line of Tunis-Carthage reported by  
 365 Zouari et al. (1985) according to the equation  $\delta^2\text{H} = 8 \delta^{18}\text{O} + 12.4$ . G1, G2 and G3 correspond to group 1, group  
 366 2 and group 3, respectively

367 The comparison of the isotopic composition ( $\delta^{18}\text{O}$ ,  $\delta^2\text{H}$ ) of groundwater between the different sampling  
 368 campaigns is useful to assess the recharge evolution and the occurrence of seasonal variations. Analytical errors

369 of isotopic measurements were considered with  $\pm 0.1\text{‰}$  vs. SMOW for  $\delta^{18}\text{O}$  and  $\pm 1\text{‰}$  vs. SMOW for  $\delta^2\text{H}$ . The  
 370 observed difference in isotopic composition is considered as not significant if variation values belong to the  
 371 margin of analytical error. Figures 11 and 12 show a small variation in isotopic signatures for all sampling  
 372 campaigns for both the shallow and the deep aquifer, except for two samples from the shallow one which are  
 373 characterized by extreme values (wells 11 and 23). For the shallow aquifer, seasonal variation of isotopic  
 374 composition is significant for samples 3, 11, 19, 23 and 26 for both  $\delta^{18}\text{O}$  and for  $\delta^2\text{H}$  contents (Fig. 11). In the  
 375 deep aquifer (Fig. 12), stable isotopes variation is significant for wells n° 103, 104, 112, 113, 114, 116 and 117.  
 376 These variations could be related to the change of climatic parameters from one season to another. These results  
 377 reveal the relationship between isotopic composition and wells' depth. In fact, the most depleted samples  
 378 represent the deeper wells whereas the most enriched samples characterize the nearest points to earth surface (i.e.  
 379 situated at depths varying between 15 and 40 m). These points are more exposed to the phenomenon of  
 380 evaporation, which depends on both atmospheric temperature and the degree of sunlight. Therefore, stable  
 381 isotope compositions of the groundwater samples vary with the depth: going deeper into the aquifer the isotopic  
 382 composition will be more depleted.

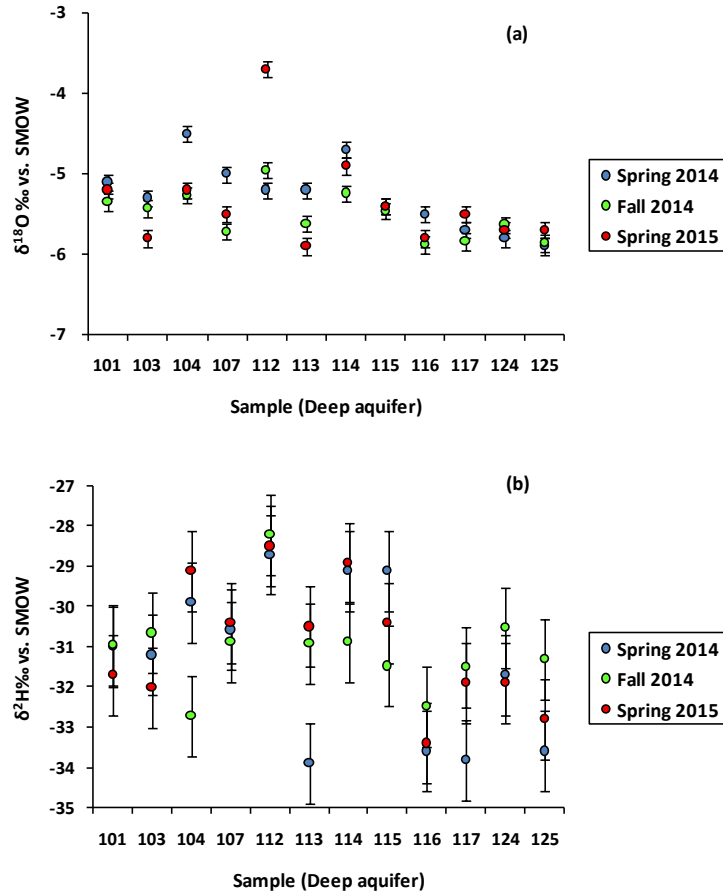
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384

385 **Fig.11** Seasonal variation of isotopic composition for shallow aquifer taking into account analytical error with: **a**  
 386  $\delta^{18}\text{O}$  values with error rate of  $\pm 0.1\text{‰}$  vs. SMOW; **b**  $\delta^2\text{H}$  contents with error rate of  $\pm 1\text{‰}$  vs. SMOW

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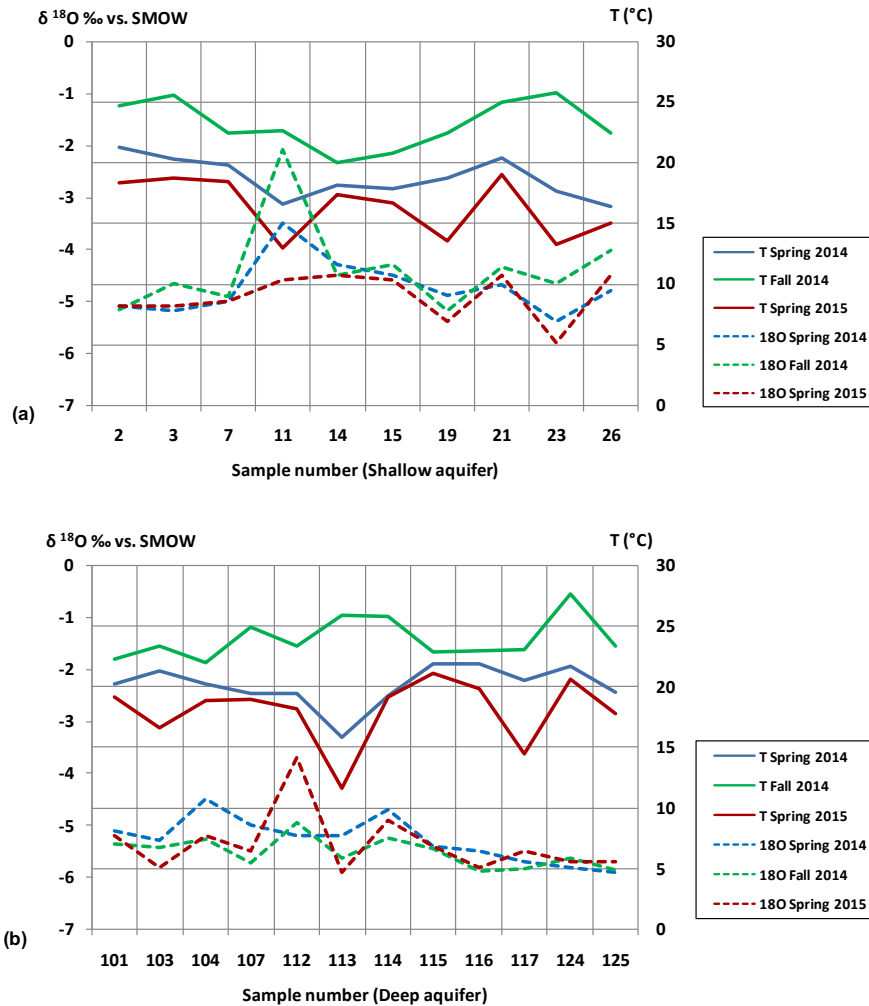


388

389 **Fig.12** Seasonal variation of isotopic composition for deep aquifer taking into account analytical error with:  
 390 **a**  $\delta^{18}\text{O}$  values with error rate of  $\pm 0.1\text{‰}$  vs. SMOW; **b**  $\delta^2\text{H}$  contents with error of  $\pm 1\text{‰}$  vs. SMOW

391 In fact, water temperature increased in dry season (fall 2014) for both aquifers, with temperature values ranging  
 392 from 20 to 25.8 °C and from 22 to 27.7 °C for shallow and deep aquifers, respectively. As shown in Fig. 13,  
 393 samples collected in fall 2014 in the deep aquifer are present the more depleted for  $\delta^{18}\text{O}$  values, compared with  
 394 the other campaigns. On the other hand, samples collected in the same period in the shallow aquifer are more  
 395 enriched, meaning that this water is more affected by evaporative process than the one collected in spring season.  
 396 This could be due to the semi-arid climate, characterizing the study area, with high variation of temperature and  
 397 precipitation intensity. Therefore, for the shallow aquifer, temporal variations of water temperature are  
 398 accompanied by a variation of  $\delta^{18}\text{O}$  contents (same order of colour for the temperature and  $\delta^{18}\text{O}$ ). However, for

399 the deep water, the temperature does not follow the same trend as  $\delta^{18}\text{O}$  contents which mean that there are other  
 400 processes that intervene other than evaporation (e.g. mixing processes and/or the variability of wells depths).  
 401

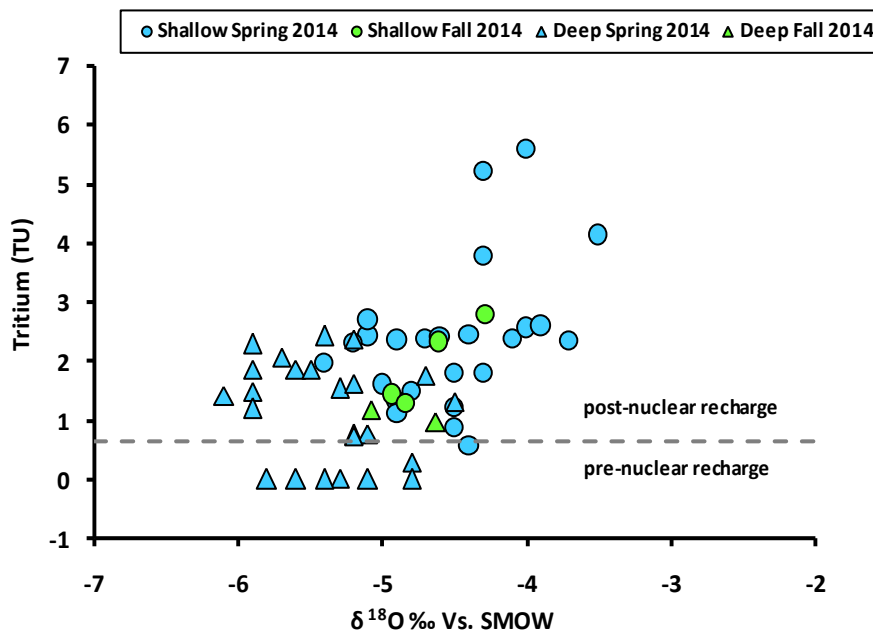


402  
 403 **Fig. 13** Seasonal variation of isotopic composition ( $\delta^{18}\text{O}$ ) and groundwater temperature ( $T$ ) for: **a** shallow  
 404 aquifer; **b** deep aquifer. *Continuous line* represented temperature line and *discontinuous line* represented  $\delta^{18}\text{O}$   
 405 values

406 Tritium ( $^3\text{H}$ ) analysis is very useful providing an indication of young water ages (Dulinski et al. 2003). Its short  
 407 half-life is about  $12.32 \pm 0.02$  years (Lucas and Unterweger 2000) allowing it to be used for identifying recent  
 408 water recharge ( $< 50$  years) according to Mann et al. (1982). Tritium contents in shallow aquifer samples vary  
 409 between 0.6 and  $5.6 \pm 0.3$  TU (Tritium Unit). Those of deep groundwater range from 0 to  $2.4 \pm 0.3$  TU for  
 410 spring 2014 period. More than 70% of water samples from deep aquifer present significant tritium values. As  
 411 most samples from both shallow and deep aquifers are characterized by values greater than 0.65 TU, it means  
 412 that most waters are post-nuclear recharged waters, i.e. groundwater recharged after the period of 1960 (Clark

413 and Fritz 1997). These high values of tritium are found almost in the south-east of the basin. This means that  
414 direct infiltration of recent rain water, characterizing by tritium content around 5.9 TU (GNIP 2007), contributes  
415 to the groundwater recharge especially at the foothills of Abderrahman Mountain. Tritium variation may be due  
416 to the elevation effect and/or the recharge during the depleted winter period as there are depleted samples in  $\delta^{18}\text{O}$   
417 (equal to -6 ‰ vs. SMOW) with significant tritium levels.

418 Few water samples display low values of tritium, less than 0.65 TU. Such the absence of tritium in some samples  
419 (Wells 103, 115, 121, 122 and 124) could be explained by either an old origin of water or the existence of mixed  
420 groundwater in the studied basin system (Fig. 14). It means that aquifer recharge was likely occurred during a  
421 cooler climate in the past. These observations are agreeing with the previous results found by Charfi et al.  
422 (2013b), which proved that recharge of deep aquifer, in some sites of the basin, was occurred during the late  
423 Pleistocene and early Holocene periods based on carbon-14 analyses. The few samples taken during fall 2014 are  
424 characterized by lower values in tritium highlighting the limited recharge at the end of summer season.



425

426

Fig. 14 Plot of tritium versus  $\delta^{18}\text{O}$

427

#### 4.4. Mixing process and quantifying groundwater flow exchange

428

429

430

Isotopic composition of shallow and deep aquifers may result from a contribution of shallow aquifer to the deep groundwater recharge. The variability of hydraulic head between these two aquifers, as shown in the piezometric map (Fig. 5), has increased by the over-exploitation of the shallow aquifer.



431 Thus, the deep aquifer would represent a mixing between old water and descending shallow groundwater, as  
 432 suggested previously by the piezometric investigation. The mixing rate between shallow and deep groundwater,  
 433 could be estimated using the isotopic mass balance (Trabelsi et al. 2009; Carreira et al. 2014; Ayadi et al. 2016)  
 434 according to the equation (5) reported by Clark and Fritz (1997):

435 
$$(X_1+X_2) \delta E_{\text{sample}} = X_1 \delta E_s + X_2 \delta E_d \quad (5)$$

436 With

437  $X_1$ : fraction of shallow aquifer;  $X_2$ : fraction of deep aquifer with  $X_1+X_2=1$ ; and  $\delta E_{\text{sample}}$ ,  $\delta E_s$  and  $\delta E_d$  representing  
 438 the isotopic composition ( $\delta^{18}\text{O}$  or  $\delta^2\text{H}$ ) of the sample, shallow aquifer and deep groundwater respectively.

439 The mixing rates were calculated by isotope mass balance using both  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  for all samples during three  
 440 campaigns. To study the contribution of shallow groundwater to the deep aquifer recharge, the mean values of  
 441 isotopic composition adopted for the shallow aquifer are  $-4.6 \pm 0.2\text{‰}$  vs. SMOW for  $\delta^{18}\text{O}$  and  $-28.5 \pm 1\text{‰}$  vs.  
 442 SMOW for  $\delta^2\text{H}$ . The mean values of  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  used for the deep aquifer are  $-5.3 \pm 0.2$  and  $-31.6 \pm 1\text{‰}$  vs.  
 443 SMOW, respectively. The results of the mixing rates are presented in Table 3. The 0% value means that there is  
 444 no contribution, whereas 100% (well 104, campaign spring 2014) represent the maximum of contribution. Fifty-  
 445 four percent of the total groundwater samples, collected during spring season in 2014, present no contribution of  
 446 shallow aquifer to deep aquifer recharge, 27% have isotopic balance (IB) rates ranging between 1 and 50% and  
 447 only 19% of samples present a high mixture rates varying between 51 and 100%. Computed values of the mixing  
 448 proportion indicate that the descending vertical leakage of the Grombalia aquifer is significant in some region  
 449 especially in north-east part of the basin where the contribution can reach 100%. According to isotope balance,  
 450 as shown in Table 3, the mixture rate is most important in spring season. This is already highlighted by both  $\delta^{18}\text{O}$   
 451 and  $\delta^2\text{H}$  isotope balances except for some samples (e.g. 103, 124 and 125).

452

453 **Table 3** Groundwater tritium contents and mixing rate (%) for the deep aquifer

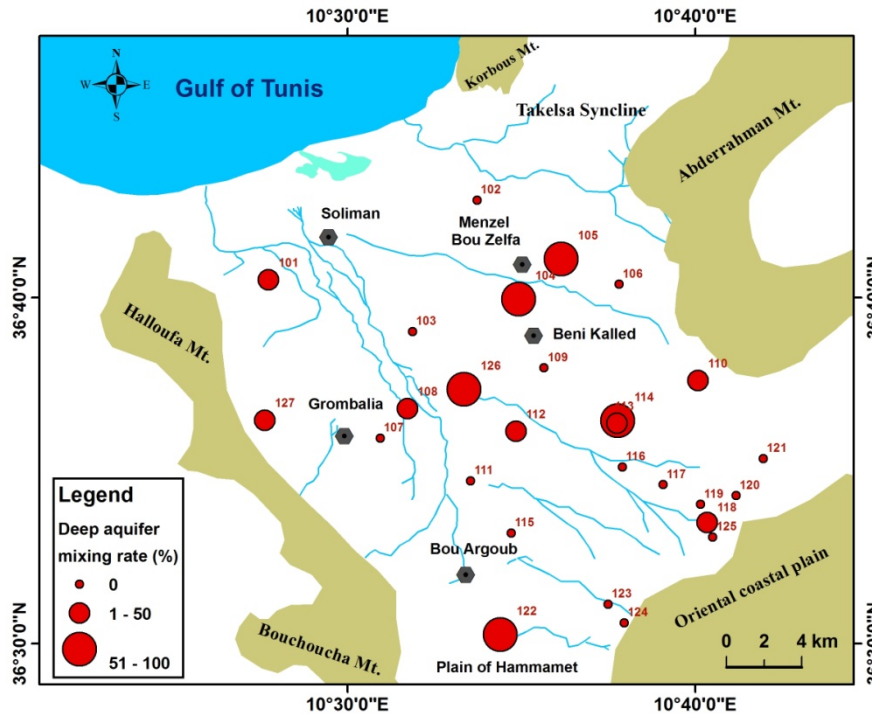
Well	$^3\text{H}$ (TU)	$^{18}\text{O}$ IB (%)	$^2\text{H}$ IB (%)	$^{18}\text{O}$ IB (%)	$^2\text{H}$ IB (%)	$^{18}\text{O}$ IB (%)	$^2\text{H}$ IB (%)
	Spring 2014	Spring 2014	Spring 2014	Fall 2014	Fall 2014	Spring 2015	Spring 2015
101	0	29	19	0	21	14	0
102	1.5	0	0	ND	ND	ND	ND
103	0	0	13	0	30	0	0
104	1.3	100	55	4	0	14	81
105	0.3	71	0	ND	ND	ND	ND
106	1.9	0	0	ND	ND	ND	ND
107	0	ND	ND	0	24	0	39

108	0.7	14	52	ND	ND	ND	ND
109	1.4	0	0	ND	ND	ND	ND
110	0.8	29	0	ND	ND	ND	ND
111	2.4	0	32	0	28	ND	ND
112	1.6	14	94	50	100	100	100
113	0.7	14	0	0	22	0	35
114	1.8	86	81	9	23	57	87
115	0	0	81	0	4	0	39
116	1.9	0	0	0	0	0	0
117	2.0	0	0	0	3	0	0
118	2.4	14	0	ND	ND	ND	ND
119	1.5	0	0	0	0	ND	ND
120	2.3	0	0	ND	ND	ND	ND
121	0	0	0	ND	ND	ND	ND
122	0	71	58	ND	ND	ND	ND
123	1.8	0	0	ND	ND	ND	ND
124	0	0	0	0	35	0	0
125	1.2	0	0	0	9	0	0
126	1.0	96	96	96	96	0	0
127	1.2	31	35	31	35	0	0

454 *IB*: isotopic balance, corresponds to the contribution of shallow aquifer to the recharge of deep groundwater, negative values  
455 are represented by 0% which means no contribution, whereas values equal to 100% expressed the maximum of contribution;  
456 *ND* expressed no data

457

458 The estimated values of the shallow groundwater contribution to the deep aquifer recharge, based on the  $\delta^{18}\text{O}$   
459 balance, appear to be the most reliable confirming findings of piezometric study as discussed previously. Figure  
460 15 shows the spatial distribution of groundwater flow exchange (%) between shallow and deep aquifer systems  
461 of Grombalia basin using  $\delta^{18}\text{O}$  mass balance. The contribution of the shallow aquifer is very limited spatially, as  
462 just a few samples show significant values. The most important mixing rates seem to occur in the north-east and  
463 the centre of the plain (south of Beni Kalled and Menzel Bou Zelfa), whereas in the south of the plain the  
464 contribution of shallow aquifer to deep aquifer recharge is very low.



465

466 **Fig. 15** Spatial distribution of groundwater flow exchange (%) between shallow and deep aquifers showing the  
 467 contribution of shallow aquifer to deep aquifer recharge using  $\delta^{18}\text{O}$  mass balance for spring season in 2014  
 468

469

### 5. Management implications

470 Hydrogeochemical and isotopic findings of the present study highlight the critical state of groundwater resources  
 471 in the Grombalia region and provide relevant information on groundwater quantity and quality: a fundamental  
 472 requisite for the implementation of sustainable management strategies. The obtained chemical data and the  
 473 associated socio-economic assessment (Re et al. 2017; Tringali et al. 2017) reveal that the uncontrolled use of  
 474 fertilizers and lack of environmental awareness are the main drivers for the increase of contaminants leaching  
 475 from surface sources to groundwater reservoirs, threatening both groundwater quality and the long-term water  
 476 supply capacity of the region. On the other hand, groundwater over-exploitation, due to the arbitrary  
 477 proliferation of pumping-wells, is the main cause of piezometric levels' decrease, as some farmers tend to dig  
 478 deeper wells when existing ones run dry.

479 The studied basin shows an example of two connected aquifer layers, where nitrate pollution affecting the  
 480 shallow aquifer continue to pollute the deeper one. Consequently, if nitrate contamination, salinization and over-  
 481 exploitation will continue with current trends, the deep aquifer would stop being considered as a strategic  
 482 groundwater reservoir for future groundwater development. Therefore, groundwater resources, and especially the  
 483 area where mixing processes are dominant, have to be protected against anthropogenic pollution and salinization,

484 by limiting groundwater abstraction and increasing artificial recharge. In fact, the adoption of preventative  
485 measures is still the most appropriate approach to protect natural water resources.

486 With regard to nitrate contents in groundwater, the best suggestion to avoid nitrate pollution is to reduce the  
487 nitrogen fertilizers application, hence collaborating with farmers and local stakeholders (Re et al. 2017; Tringali  
488 et al. 2017), and to implement a drainage system to collect irrigated water, instead of allowing it draining into a  
489 the *wadis* or leaching to groundwater.

490 During the last decades, water shortage become an increasing problem derived from several causes such as land  
491 use change, climate variability, pollution and diversion of surface water (Pereira et al. 2009). As consequence,  
492 water deficit caused a significant deterioration of agricultural activities, resulting in land abandonment,  
493 particularly in coastal areas. This led the public authorities in Grombalia region to implement three dams,  
494 namely the Bezigh (built in 1954, with capacity of 6.5 Mm<sup>3</sup>/year) situated upstream of Menzel Bou Zelfa region,  
495 the El Masri (built in 1968, with capacity of 6.9 Mm<sup>3</sup>/year) and Tahouna dams (built in 1967, with capacity of  
496 0.96 Mm<sup>3</sup>/year) situated upstream of Bou Argoub region (Lachaal et al. 2016). Also, since the end of the 1980s  
497 and onwards, a vast program of water resources mobilization was implemented targeted to the safeguard of  
498 irrigated agriculture and the supply of drinking water to urban areas in the region. Thus, the Medjerda-Cap Bon  
499 canal was set up to ensure the transfer of water from the north collected by several dams built on the country's  
500 longest river Medjerda to several irrigated perimeters in the Grombalia region. The transferred water,  
501 characterized by an average salinity of 1000 mg/L (DGRE 2015), is used for irrigation and for artificial  
502 groundwater recharge in some sites since 1992. Indeed, despite of the fact that many management plans have  
503 been applied in Grombalia region since 1974, such as adopting artificial recharge by water injection in shallow  
504 wells and infiltration basins building, water supply stress is still the main concern faced by this agricultural  
505 coastal area. In addition, recent climate change coupled to population growth and the associated rise in water and  
506 food demand, are contributing increasing the gap between demand and available water resources, and may harm  
507 groundwater resources availability in the long-run (especially when the drawdown affects the deep aquifer  
508 system).

509 Results of the present investigation hence reveal the weakness of current groundwater management in the region,  
510 due to (i) a poor control and regulation on private wells and their abstraction; (ii) an often inequitable access to  
511 water resources; and (iii) the lack of sewage treatment network. In addition, as a consequence of the scarce  
512 availability of water resources, the agricultural base in this region fails to provide employment to the entire  
513 population located in the rural areas. This generates an internal migration from rural to urban areas, which not

514 only contributes hampering the issue of land abandonment, but also generates overcrowding of cities by non-  
515 skilled labour force seeking jobs in different areas of employment (e.g. trade, industry, transport and services).  
516 This situation, common to many arid and semi-arid regions worldwide, including the MENA countries, is  
517 increasingly becoming source of social instability and often the driver for riots and conflicts at both local and  
518 regional level (Robins and Fergusson 2014; Miletto et al. 2017). Therefore, since business as usual management  
519 actions, focused on dams and reservoirs constructions, did not prove to be effective in mitigating the effects of  
520 piezometric level decrease, it will be of paramount importance to seek for alternative strategies to bridge the  
521 supply-demand gap, by focusing on both scientific innovation and participative actions (Kinzelbach et al. 2003).  
522 This means to increase the investigations and associated investments on managed aquifer recharge, irrigation and  
523 crops' yield optimization, but also to collaborate with local population and water regulators in order to promote  
524 the implementation of effective water conservation strategies and contamination source control. The latter could  
525 be fostered by hydrogeologists, as providers of sound information on groundwater status that can also be actively  
526 engaged in public participation of local stakeholders and bottom-up investigations targeted to rural development,  
527 according to the socio-hydrogeological approach proposed by Re (2015). Indeed, sustainable groundwater  
528 management cannot be achieved in the long-term without a sound knowledge of the hydrogeochemical  
529 properties of water resources and this study also reveals the fundamental role of multidisciplinary investigations  
530 to support the sustainable development in arid and semi-arid regions with the potential to reduce poverty in rural  
531 areas.

532

### 533 **Conclusion**

534 Hydrochemical and isotopic investigations performed in Grombalia aquifer system (NE Tunisia) permitted to  
535 better define groundwater chemical composition and to preliminary assess the evolution of groundwater quality  
536 according to seasonal variations. Chemical data showed high salinity of the analysed groundwater samples with  
537 significant variation in time and space. Several factors are controlling the groundwater quality traducing the  
538 complexity and the interference of several geochemical processes. The first process expresses the phenomenon  
539 of water-rock interactions which is enhanced with longer residence time, the second represents the evaporation  
540 process affecting mainly the shallow groundwater and return flow of evaporated irrigation water, the third  
541 process represents cation exchange reactions with clay minerals and the fourth one is expressed by mixture  
542 process between shallow and deep aquifers. The present study reveals also that anthropogenic factor contributes  
543 effectively to groundwater mineralization. Nitrate concentrations achieved in many places already alerting levels

544 exceeding the drinking water limit (50 mg/L). Determination of agricultural parameters coupled with high total  
545 hardness and electrical conductivity values indicate the unsuitability of most groundwater samples for drinking  
546 and irrigation purposes. The spatial distribution of SAR values, using ordinary kriging technique reveals that  
547 high values occurred most in the north of the region near the coast. Concerning isotopic composition,  
548 groundwater displays large range of variation. The stable isotope composition of groundwater samples and  
549 tritium contents proved the existence of three groundwater groups. The first one is characterized by a strong  
550 contribution of modern precipitation to aquifer recharge, confirming the recent recharge by rapid infiltration of  
551 rain water indicate either recent origin or the existence of mixing process. However, the second group  
552 corresponds to the most enriched samples dominated by an evaporation effect. The third group represents the  
553 most depleted groundwater samples from deep aquifer that is probably recharged during cooler climate.  
554 Therefore, the fact that these two aquifers are characterized by a similar isotopic signature represents a sign of  
555 water exchange between these two aquifers. The comparison of groundwater isotopic composition under  
556 seasons, taken into account the analytical errors, reveals that seasonal variations affect stable isotope  
557 compositions especially for the shallower wells. Isotopic mass balance, using both  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  isotopes, has  
558 confirmed the mixture between shallow and deep aquifers in some sites of the basin and allowed to quantify the  
559 mixing percent as a preliminary synthesis of mixing rates between the different aquifers of Grombalia. The over-  
560 exploitation state of groundwater resources needs to be properly managed as part of integrated plans for the  
561 basin's future supplies. The scientific results of this study have important implications for groundwater  
562 management in the Grombalia basin and represent an important contribution to the overall assessment of  
563 groundwater quality in the MENA region. This work highlights the need for implementing new measures to  
564 bridge the gap between water supply and demand, with concerted efforts by various agencies and local (private  
565 and public) stakeholders. In addition, as concerns the role of hydrogeology and hydrogeochemistry in supporting  
566 aquifer protection, special attention should be dedicated to the realization of a permanent monitoring network,  
567 targeted to the control and mitigation of aquifer salinization and over-exploitation. Seasonal monitoring data  
568 would be an asset for the development of numerical groundwater flow models, useful to predict aquifer  
569 behaviours under different climatic and recharge scenarios.

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