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Nitrate contamination of Sminja aquifer groundwater in Zaghouan, northeast Tunisia: WQI and GIS assessments

Meriem Ameur*, Fadoua Hamzaoui–Azaza, Moncef Gueddari

Research Unit of Geochemistry and Environmental Geology, Faculty of Science of Tunis, University of Tunis El Manar, 2092 University campus Farhat Hached, Tunis, Tunisia, emails: meriem_ameur@yahoo.fr (M. Ameur), Fadoua_fst@yahoo.fr (F. Hamzaoui–Azaza), Moncef_gueddari@yahoo.fr (M. Gueddari)

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ABSTRACT

Groundwater resources play a very important role in satisfying people's everyday needs in terms of consumption, irrigation and tourism. In Tunisia, as in most countries in the Mediterranean, groundwaters are fragile, under an arid or semi-arid climate, vulnerable to pollution, and influenced by climate change. The Sminja aquifer in northeast Tunisia is one of the major sources of fresh water for the city of Zaghouan. Twenty-three wells and boreholes were sampled in January 2013 and subjected to salinity analysis of major elements and nitrates. To study the geochemical quality of the Sminja aquifer, we used a geographic information system to develop geographic location maps and spatial distribution maps of nitrates, salinity, and the water quality index (WQI) of the study area. Nitrate concentrations ranged between 8 and 137 mg/l. The highest values of this parameter in groundwater samples found in northern and eastern parts of the Sminja aquifer have a direct relation with the fertilization of agricultural land. The salinity of the water varies from 1,170 upstream to 9,570 mg/l downstream of the aquifer. The parameters that were considered in the calculation of the WQI were calcium, magnesium bicarbonate, sodium, chloride, nitrate, sulfate, potassium, and total dissolved salts. Based on the values of the WQI, the water samples of Sminja aquifer were classified into three groups: poor water quality, very poor water quality, and unfit for consumption (the FS1 well). The groundwaters of the Sminja aquifer, apart from the southern part, do not meet the drinking water standards set by the World Health Organization with respect to salinity and nitrate levels.

Keywords: GIS; Hydrogeochemistry; Nitrates; Salinity; Sminja aquifer; Tunisia

1. Introduction

Climatic conditions, demographic growth and the unequal distribution of water in space and time increase water demand and these constraints threaten water resources in Tunisia [1,2]. The concentration of nitrates in drinking water can be an indicator of water quality. The scarcity and the deterioration of water resources are also the main factors that hinder the socioeconomic development of the country. The Sminja aquifer is exploited for irrigation and to satisfy the drinking water needs of the Bir Hlima, Mograne, and Sminja regions. The limit of nitrate content in water is set by the World Health Organization (WHO)

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*Corresponding author.

at 50 mg/l. High concentrations of nitrates can cause serious health problems such as methaemoglobinaemia or “blue baby” syndrome [3].

2. Study area

The Sminja aquifer is located in the Zaghouan governorate (the north-eastern part of Tunisia) and extends geographically between 9.95° and 10.09° East Longitude and from 36.37° to 36.46° North latitude (Fig. 1).

With a total area of 2,820 km^2 (1.79% of the area of Tunisia), the governorate is crossed by the Tunisian dorsal characterized by the domination of high reliefs, especially the mountain of Zaghouan with an altitude of 1,295 m which constitutes the highest point in the region [4]. The climate of the study area is upper semi-arid; the average annual temperature in the Zaghouan station is around 18°C, and the mean annual rainfall is about 500 mm [5].

The basin of Oued Meliane is the principal drainage basin of the Sminja aquifer. The main wadis which feed the aquifer are the Meliane, Sidi Hmida, Naoura, Melah, and Saad.

The soils of the area can be classified into five types; raw mineral soils, poorly developed soils, calcimorphic soils, isohumic soil, and vertisoils [6].

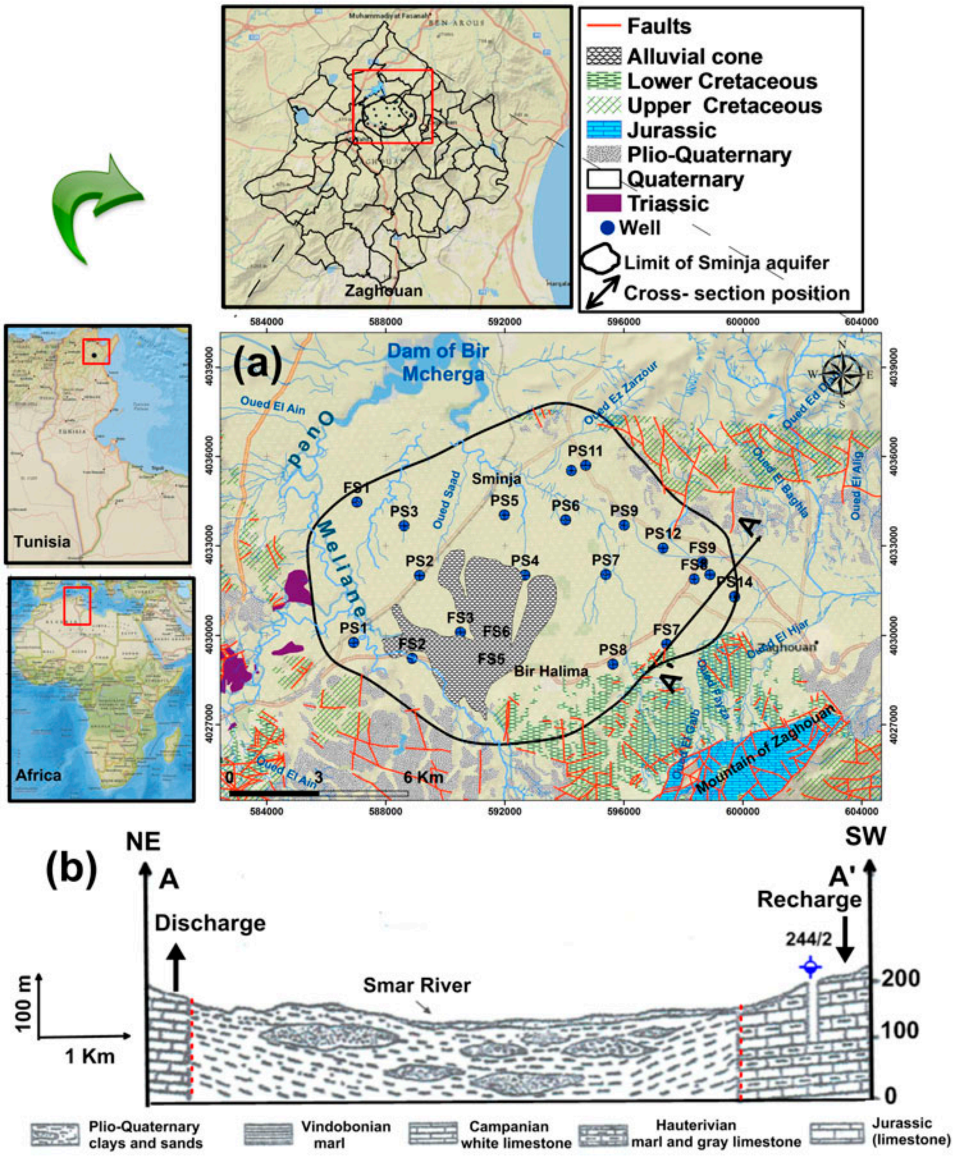

Fig. 1. Geologic map: (a) and schematic hydrogeological cross section (b) showing the hydrodynamic of the Sminja aquifer.

Agricultural activity in the Zaghouan region is mainly determined by the semi-arid climatic conditions and the quality and quantity of groundwater resources and it is based on cereals and arboriculture (mainly olives).

The region has a large agricultural area of about 282.000 ha of which 185.000 ha are cultivable land and 87.000 ha are pasture and forests. Cereal production averages 1.4 million quintals (UK = hundredweight) [7].

In Sminja aquifer, the groundwater flow originates mainly from the South of Sminja and the northwest of Bir Mcherga. Transmissivity is calculated from flow rate test made on many boreholes (Bir Halima from 1 to 5 and Jabbes) and we obtained the following value $7.3 \times 10^{-3}\ \mathrm{m}^2/\mathrm{s}$. This value corresponds to a more permeable zone than other areas of the Sminja region where the transmissivity is much lower than the value of $4.5 \times 10^{-3}\ \mathrm{m}^2/\mathrm{s}$. Based on exploratory drilling data and the electrical prospecting campaign, then we simply take the value of $10^{-3}\ \mathrm{m}^2/\mathrm{s}$ as the average value of all Sminja water samples. The groundwater flow rate of the water aquifer would be 60 l/s, which means 1.9 million m^3/year [8]. The average depth of water sources in Sminja shallow aquifer is about 90 m.

3. Geology and hydrogeology

The study area is a vast plain covered with Quaternary sediments clay, sand and gravel and bordered to the south by a Djebels area (Mio-Pliocene and Cretaceous) [9].

The formations encountered in our study area are staged Triassic to Quaternary. The study area represents the joining of two geological maps at a scale of 1: 50.000, which are Zaghouan and Bouficha.

The Sminja region is a collapse plain separated by reliefs that surround it by more or less visible tectonic accidents. Outcrops are mainly the Jurassic, Cretaceous, Tertiary, and Quaternary. The Triassic is visible as a diapiric form.

In connection with the tectonics, the Triassic appears as small islands at the anticlinal axes. It is found in Zaghouan, Djebel Mcella, and Hammam Djedidi. The Triassic is rarely in normal position [10]. The Triassic outcrops mostly punctuate abnormal contacts [11].

The Lower Jurassic (Lias) is present in Djebels Zaghouan, El Ouest, El Aziz, Beni Kleb, Rouass, and Rouissat. It is formed almost exclusively by zoogenic, oolitic, or sublithographic limestone. The Upper Jurassic is represented by calcareous series with small marl intercalations.

Lower Cretaceous sediments are found in the mountains of Zaghouan, near Djebels Kleb, Rouass, and Rouissat, with the city of El Fahs and Djebels Ouest and Maouine.

The Lower Cretaceous is represented by alternations of marl and limestone except Aptian that is formed by clays and sandstones. The Upper Cretaceous is represented by limestone and marl. It is visible on the sides of Mio-Pliocene syncline between Djebel Zaghouan and Pont d’El Fahs and in center of the anticline Djebel Jahfa. Tertiary sedimentation starts with marl returnable to Paleocene formation of El Haria. Then, Eocene is represented by the limestone formations Metlaoui to the base and marls and Souar clays at the top.

The outcrops of the Oligocene appear at Djebel Jahfa and south of Djebel El Ouest. These outcrops are made from bottom to top by the alternations of clays and friable sandstone and finally coarse sandstone. The Burdigalian is the Ain Ghrab formation, which outcrops at Djebel Jahfa. The Vindobonian is present in the South and West of Djebel Jahfa. The set begins with sandy loams and ends with white and reddish sandstone.

The Pliocene–Quaternary is composed of conglomerate alternations, sandstone, and clay and known to the west of Djebel Jahfa and between Djebel Zaghouan, and the town of El Fahs. The Quaternary formations are widely represented either in the plains or in the foothills.

Section A-A´ (Fig. 1(b)) oriented NE–SW and passing from the right side of the drilling (244/2). It shows that the Pliocene–Quaternary filling is reduced to the Oued Smar Valley. The discontinuity appeared in the facies result of the system of NW–SE faults that puts contact Pliocene–Quaternary and Lower Cretaceous (Hauterivian). Drilling (244/2) crossed over 106 m deep a set formed by repeated alternations of calcareous marl and limestone beds [12].

4. Methodology

4.1. Spatial analysis of groundwater quality

Twenty-three groundwater samples were collected in polyethylene bottles from active pumping wells belonging to Sminja aquifer in January 2013. Standard methods (Table 1) were used for physicochemical analysis of nutrient elements. Water samples were filtered with 0.45 μm Millipore filter paper, acidified with nitric acid and stored at 4˚C.

Table 1
Methods used for nutrient elements, physicochemical ion analysis of groundwater samples [15]

Parameters	Methods
TDS, pH, *T* (℃)	Portable field kit
Cl	Mohr method ($AgNO_3$)
HCO_3	Potentiometric method
SO_4	Gravimeter method using BaCl
Nutrient elements (NO_3, NO_2, NH_4)	Colorimetric method
Ca, Mg, Na, K	Spectrometry of atomic absorption

Validation of chemical analysis results was conducted by the ionic balance verification and by repeating the analysis for the same sample. The accuracy of the results is evaluated by calculating the charge balance error (CBE). The test results are considered reliable only when the CBE is less than or equal to 5% [13]. The WHO has established guidelines and standards for drinking water quality to protect human health [14].

4.2. GIS analysis

Geographic information systems (GIS) combined with other geochemistry software can provide complete information in real time and space about the quality of water resources, their spatial distribution, and their use through the development of thematic maps: geological maps, soil maps, nitrates spatial distribution, and water quality index (WQI) of the study area (Fig. 2).

The location of sampling boreholes was determined with global positioning system (GPS).

The diagram (Fig. 2) reports the methodology adopted for the studied sites map development.

GPS Track Maker is a localization tool allowing the transfer of GPS points of a ground and recording them in GIS or Google Earth format to finally arrive at a map with sampling sites whose coordinates are previously recorded by the GPS.

With GIS, researchers can map, model, search, and analyze large amounts of collected data in a single database. In several countries, the GIS tool is widely used by government, industries, and researchers for a range of applications [16].

Among the main uses is the analysis of environmental resources including water for irrigation. In this context, the use of this tool can manage well, view, update data sampling points, make thematic maps, and cross qualities data with other data layers.

The different water quality maps were produced using GIS software, ArcMap, showing the spatial variation of the concentrations of groundwater parameters at various locations in the study area.

The GIS now present some of the best techniques for decision support by their large cartographic capabilities, spatial analysis of natural phenomena and interpretation [17].

4.3. Water quality index

The WQI is an important parameter that allows evaluation of water quality intended for human consumption [18].

5. Results and discussion

5.1. Origins of groundwater mineralization of Sminja aquifer

The underground flow of the aquifer are from the South in Sminja area and from northwest in the region of Bir Mcherga. The aquifer of Oued Rmal located to

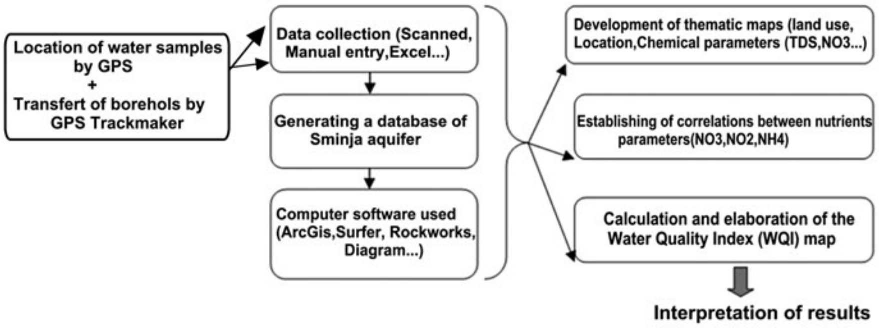

Fig. 2. Chart of the working method adopted.

the east, is the natural discharge of Sminja aquifer which in turn is diverse in the sea (Gulf of Hammamet).

Sminja aquifer is enclosed in a very heterogeneous aquifer composed of alternating sand and clay belonging to the Quaternary. The southern part of the aquifer is much more productive and provides waters with salinity less than 3 g/l. Salinity in the water samples of Sminja aquifer ranges from 1,170 to 9,570 mg/l.

The spatial distribution (Fig. 3) of these values shows that the water collected in the upstream portion of the aquifer is weakly to moderately mineralized, with a salinity between 1,000 and 3,000 mg/l, whereas those sampled in the downstream portion are most mineralized, with a salt content greater than 3,000 mg/l.

This increase in aquifer flow direction is mainly influenced by the origin of the water, the water–rock interactions, residence time and lithology, and the particle size of the unsaturated zone.

The lowest salinity values characterize the south-eastern part of the aquifer, which corresponds to the area where the maximum charging is done through the wadis.

The highest salinities characterize water taken from the northern and western parts of the aquifer, where the lithology of the aquifer is thin (mostly clay) with some outcrops of Triassic evaporate type such as gypsum from the Meliane wadi.

For all Sminja aquifer waters, concentrations of Na^+ and of Cl^- are highly and positively correlated (Fig. 4), with an around 0.87 correlation coefficient. This indicates that the main source of these ions is the dissolution of the halite .The values of $Na^+/(Na^+ + Cl^-)$, which are around 0.5 confirm this origin [19]. According to the correlation diagram of the levels of Na^+ and of Cl^-, there are two groups of water:

(1) The water having concentrations of Na^+ and of Cl^- below 60 mmol/l, representing 96% of all water sources, relates to the South and Central portions of the aquifer.
(2) Waters of FS1 drilling with concentrations of Na^+ and of Cl^- higher than the first group and above 120 mmol/l, located in the western part of the aquifer.

To identify the major elements whose concentrations control the total mineralization of the aquifer waters, we have established concentration–salinity diagrams.

The relationship between concentrations of major elements and total mineralization (Fig. 5) shows that the concentrations of Cl^-, Na^+, and SO_4^{2-} are well

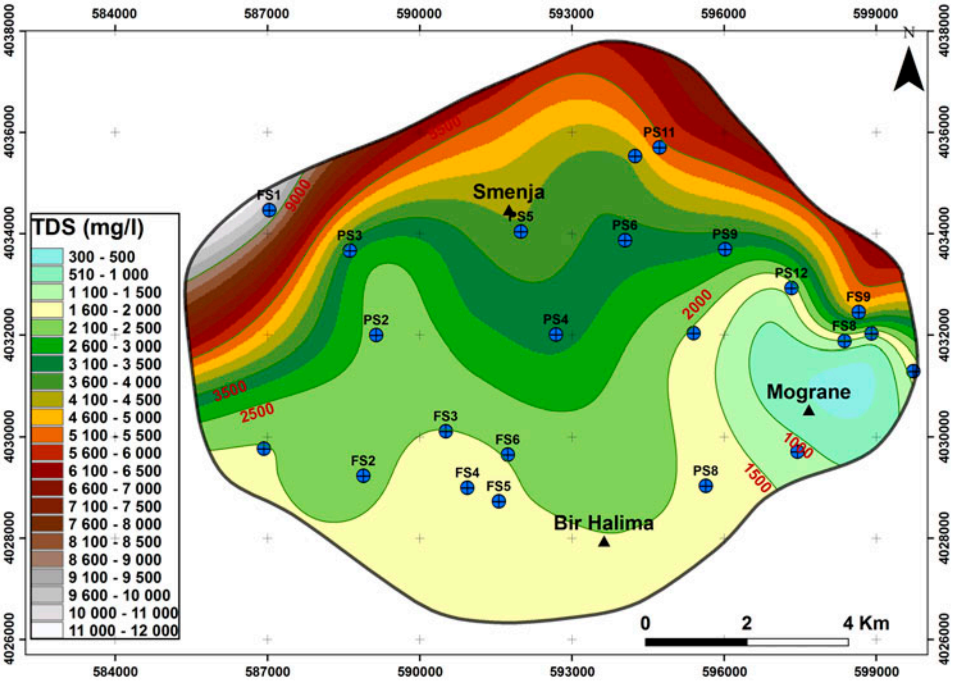

Fig. 3. Spatial distribution map of salinity (January 2013).

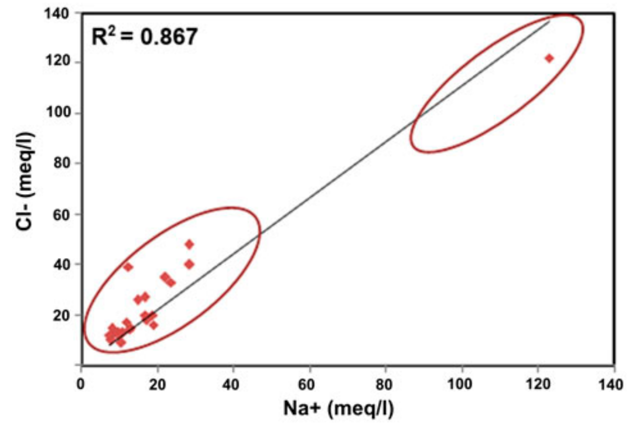

Fig. 4. Plot of Cl^- (meq/l) against Na^+ (meq/l).

correlated with salinity, with correlation coefficients, respectively of 0.90, 0.71, and 0.6 which indicates that the total salt content is mainly controlled by the concentrations of these elements. The concentrations of HCO_3^- and K^+ are not correlated with salinity.

5.2. Spatial distribution maps of nitrates

Nitrate concentrations range between 8 and 137 mg/l. The highest values that characterize the waters of the northern and eastern parts of the aquifer relate to the fertilization of agricultural land. The distribution of NO_3 concentration is illustrated in Fig. 6. Nitrate is a very important parameter for assessing the contamination of groundwater [20].

This parameter respects the desirable limit of 50 mg/l in only four samples (Table 2). Nine percent of the water samples are within the Maximum Permissible Limit.

In the study area, 74% of the water samples exceed the nitrate concentration limit (50 mg/l). All the samples have nitrite values within the desirable limit.

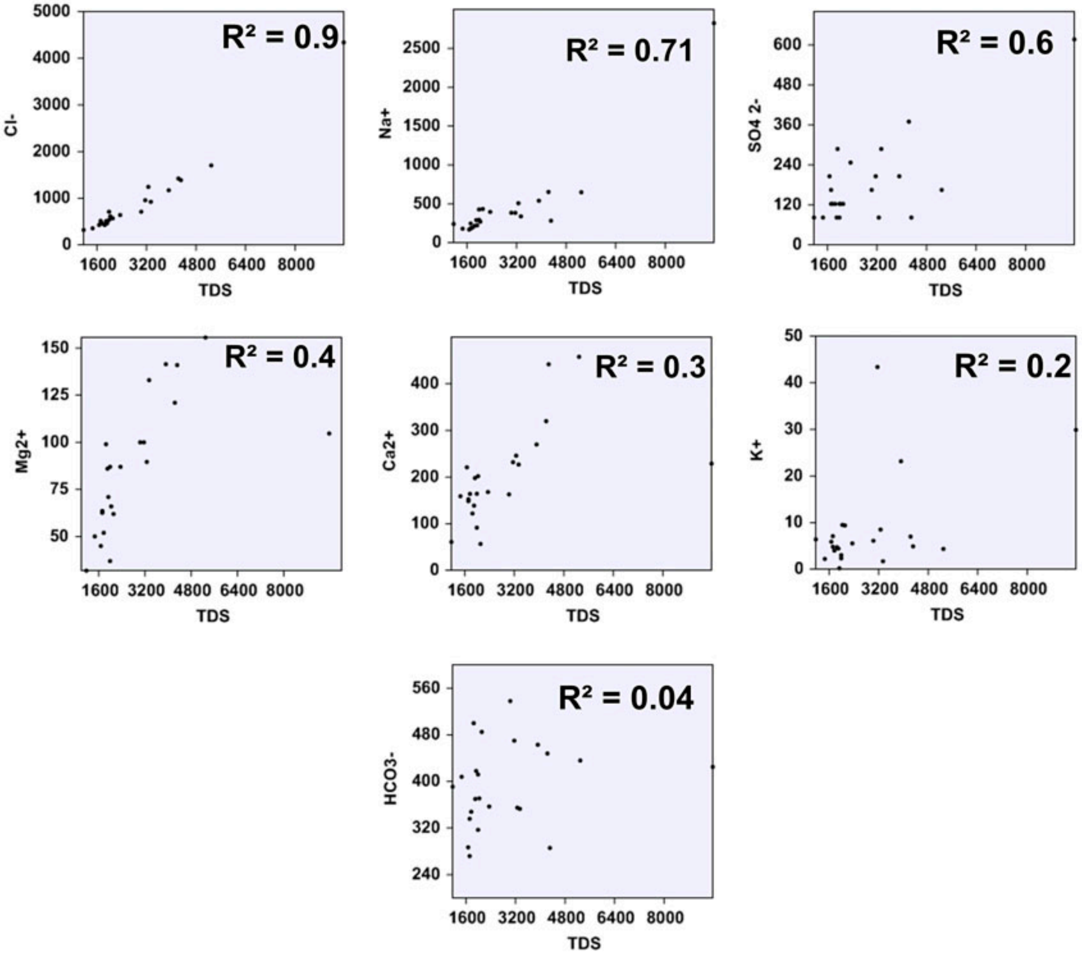

Fig. 5. Plots showing concentrations of major ions (mg/l) of Sminja aquifer versus total dissolved salts (TDS).

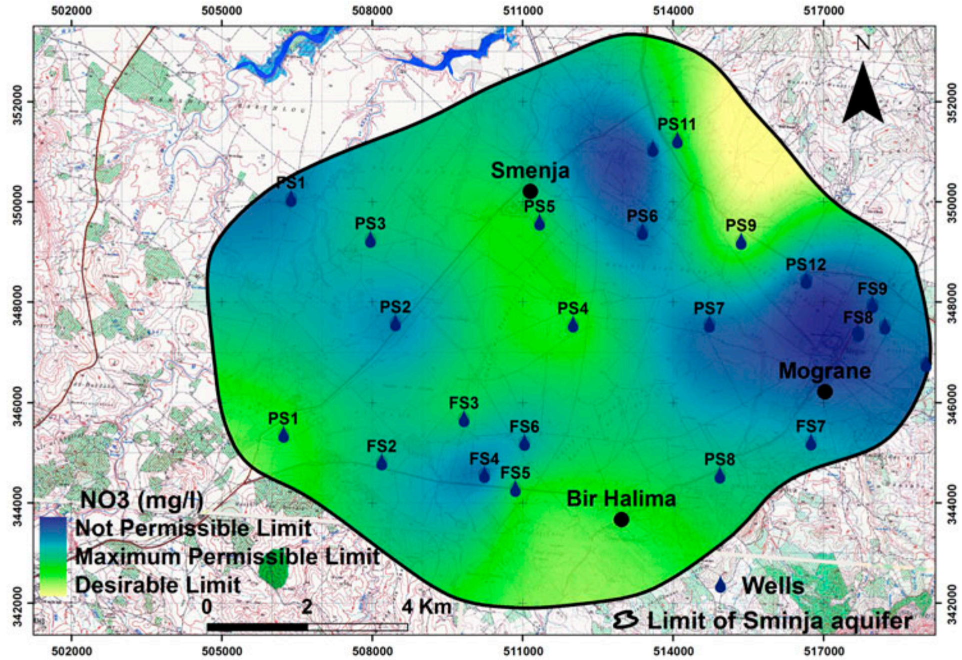

Fig. 6. Spatial distribution of nitrate (mg/l) and permissible limits.

Table 2

WHO limits on TDS, NO_3^- and NO_2^- of water samples of Sminja aquifer

Parameters (mg/l)	Desirable limit (DL)			Maximum permissible limit (MPL)			Not permissible limit (NPL)			Range parameters	
	Values	No. of samples	%	Value	No.	%	Value	No.	%	Min	Max
TDS	<100	0	0	=100	1	4	>1,000	22	96	1,117	9,570
NO_3^-	<50	4	17	=50	2	9	>50	17	74	8	137
NO_2^-	<3	23	100	=3	0	0	>3	0	0	0.05	0.08

5.3. The origin of pollution caused by nitrates in the sminja aquifer

Nitrate is the most oxygenated form of nitrogen and it is present naturally in water at levels lower than 10 mg/l [21]. Farmers use large quantities of nitrogen fertilisers, pesticides and slurry to fertilize agricultural land. These fertilizers generate high nitrate concentrations in groundwater, exceeding drinking water standards. The nitrates overloaded waters are mostly abundant in areas of intense agricultural activity belonging to shallow aquifer than for deep aquifers.The positive correlation between NO_3 and SO_4 suggests that N and S were used in the study area as fertilizers with the chemical formula $(NH_4)_2SO_4$ (Fig. 7) [21,22]. Fig. 7 shows that NO_3 and Ca are well correlated. These two elements derive mainly from overuse of fertilizers with the chemical formula $Ca\ (NO_3)_2$ [23].

5.4. Health effects of nitrate in drinking water

Nitrates are highly soluble in water and easily migrate downward to join groundwater when concentrations exceed crop needs [24].

Nitrate values greater than 50 mg/l exceed the levels recommended by the WHO. The contamination of water by nitrates causes acute methemoglobinemia in infants less than six months and pregnant women and can result in digestive disorders [25].

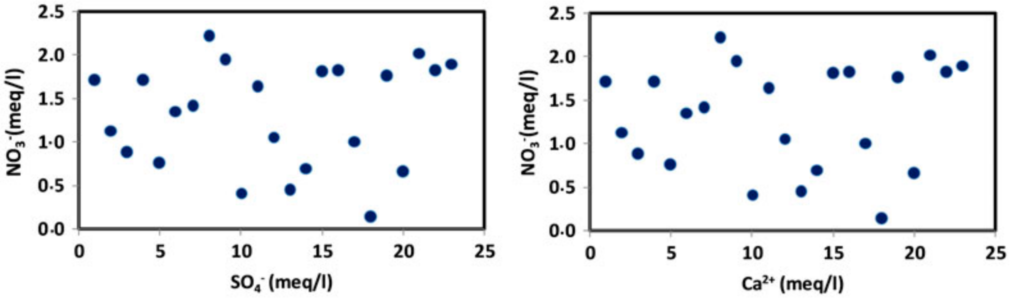

Fig. 7. Plot of NO_3 vs. SO_4 (meq/l) and Ca (meq/l).

5.5. Estimation and mapping of WQI

The WQI is a principal parameter for evaluating the potability of surface and groundwaters to freshwater supplies [26]. Horton [27] tested 10 parameters that identify water quality. In order to calculate this index, the weight wi is designated a weight to each parameter depending on its relative importance to evaluate the water quality of the Sminja aquifer (Table 3). The wi values range from one to five. The minimum wi (1) is given to potassium because it is not very harmful. The maximum weight (5) is assigned to the salinity and nitrate content, because of their magnitude in the evaluation of the water quality [28]. The second step is to calculate the relative weight (Wi) from Eq. (1):

$$\mathrm{Wi} = \mathrm{wi}/\sum ni = 1 \quad (1)$$

where n is the number of parameters, wi is the weight of each parameter.

In the third step, an evaluation scale of the quality (qi) of each parameter is calculated from Eq. (2):

Table 3
Weight and the relative weight of parameters used to calculate the WQI of the Sminja aquifer

Parameters (mg/l)	WHO (2011)	Weight (wi)	Relative weight Wi
Salinité	1,000	5	0.172
pH	8.5	3	0.103
Na^+	200	3	0.103
K^+	30	1	0.034
Ca^{2+}	200	2	0.069
Mg^{2+}	150	2	0.069
Cl^-	250	3	0.103
SO_4^{2-}	250	3	0.103
HCO_3^-	380	2	0.069
NO_3	50	5	0.172
Somme		29	

Table 4
Detailed calculation of the WQI of the borehole FS1

	Parameters (mg/l)	pH	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	NO_3	TDS
	WHO	8.5	200	30	200	150	250	250	380	50	1,000
	FS1	7.04	2,825	30	229	105	4,341	617	425	106	9,570
Eq. (1)	Wi	0.103	0.103	0.034	0.069	0.069	0.103	0.103	0.069	0.172	0.172
Eq. (2)	qi	0.82	1,412.5	99.67	114.5	52.35	1,736.4	246.8	112	211.29	296
Eq. (3)	Sli	8.6	145.49	10.30	11.79	5.39	179.63	28.47	8.59	36.43	51.03
Eq. (4)	WQI		477.93								

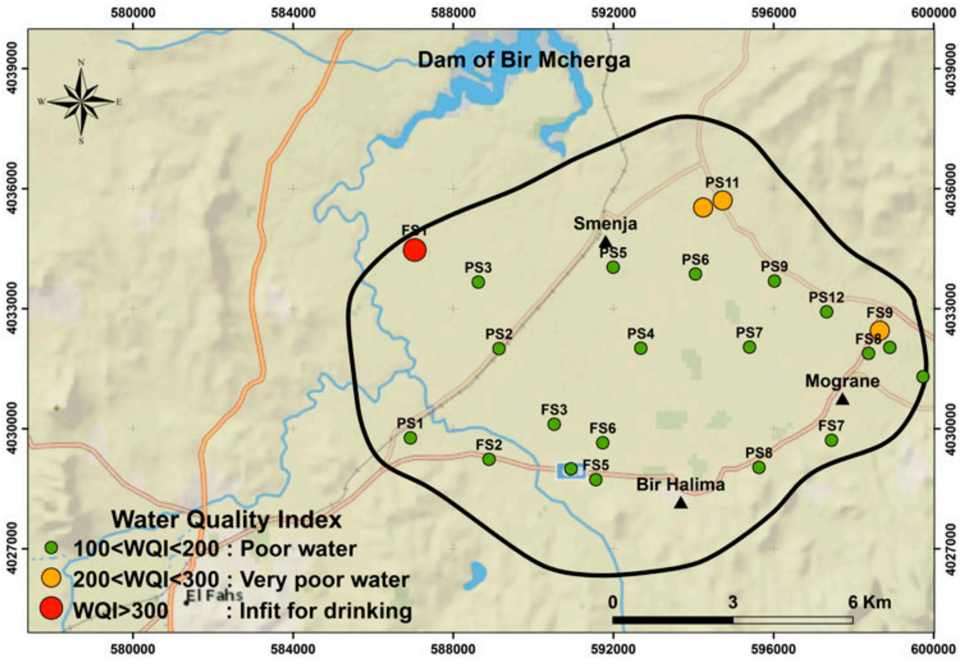

Fig. 8. Spatial distribution of WQI of Sminja aquifer.

$$qi = (Ci/Si) \times 100 \tag{2}$$

where Ci is the concentration of a component i in each sample of water, mg/l and Si: maximum permissible concentration determined by the standard of the WHO.

The index is sub SIi calculated from Eqs. (3) and (4) (Table 4):

$$SIi = Wi \times qi \tag{3}$$

Table 5
Classification of the water quality of Sminja aquifer based on the values of WQI

WQI Class	Type of water	% of water samples
<50	Excellent water	0
50–100	Good water	0
100–200	Poor water	83
200–300	Very poor water	13
>300	Unfit for drinking	4

Table 6
Water quality classification based on WQI and the % of water samples of Sminja aquifer

Samples	WQI values	Depth (m)	Classification	Samples	WQI values	Depth (m)	Classification
FS1	477.93	89	Unfit for drinking	PS4	171.64	91	Poor water
FS2	146.75	52	Poor water	PS5	191.06	60	Poor water
FS3	135.86	92	Poor water	PS6	185.73	103	Poor water
FS4	148.16	122	Poor water	PS7	156.98	98	Poor water
FS5	128.46	136	Poor water	PS8	130.97	56	Poor water
FS6	150.25	90	Poor water	PS9	138.07	120	Poor water
FS7	126.19	114	Poor water	PS10	233.22	118	Very Poor water
FS8	146.63	106	Poor water	PS11	219.67	130	Very Poor water
FS9	206.57	110	Very Poor water	PS12	149.34	102	Poor water
PS1	129.70	66	Poor water	PS13	148.09	107	Poor water
PS2	167.27	53	Poor water	PS14	147.00	109	Poor water
PS2	183.22	56	Poor water				

$$\mathrm{WQI} = \sum \mathrm{SIi} \quad (4)$$

In the Sminja aquifer, the values of WQI vary from 126 to 477 (Fig. 8). Therefore, they can be classified into three categories: "Poor water," "Very poor water," and "Unsuitable for drinking." (Table 5). Table 6 shows that 83% of water samples are of poor quality, 13% are very poor quality, and only 4% are unfit for human consumption (borehole FS1).

6. Conclusion

Tunisia has promoted irrigated agriculture to increase the production and diversity of cropping systems. Unfortunately, the irrigation water quality is in some cases misnomer. For this reason, the development of irrigation was accompanied, in most situations, by the appearance of salinization process and nitrates.

The water samples of the Sminja aquifer, except that of the southern part (FS7 and FS8), do not comply with the drinking water standards set by the WHO in respect of salinity which is in the order of 1,500 mg/l. Salinity is mainly controlled by concentrations of Na, Cl, and HCO_3, and increases from upstream to downstream, which explains the higher salinity registered in the eastern and western parts of the aquifer.

Some 78% of the water of the Sminja aquifer has nitrate levels over the limit set by the WHO (50 mg/l), with values reaching 137 mg/l. High nitrate concentrations in water cause haemoglobinaemia, inter alia.

Based on the values of the WQI, the waters of the Sminja were classified into three groups: 83% poor quality, with a range of 100 < WQI < 200, 3% very poor quality with a range of 200 < WQI < 300, and the FS1 borehole, which was described as unfit for consumption.

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