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Characterization of Groundwater and Distribution of Fluoride in the Eastern Region of the Algerian Northern Sahara (Ouargla)

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ABSTRACT

In the eastern region of the Algerian Northern Sahara, the groundwater is the only resource for drinking water supply and irrigation. This study aimed to assess the physical-chemical quality of groundwater with exposition of the fluoride distribution in the eastern region of Algeria taking as case study Ouargla area. The sampling campaign was carried out in such a way to cover the exploited aquifers (Miopliocene and Senonian). Water temperature, pH, conductivity, hardness, alkalinity, principal ions (Sodium, Potassium Calcium, Magnesium, Bicarbonates, Nitrates, Sulfates, and Chlorides) and the fluoride content in the groundwater were measured and determined. Examination and validation of obtained results were by the use ionic balance method and the hydrochemical analysis by Piper, Stabler and Richards diagrams. The obtained results of our study show that the groundwater of the Ouargla area presents a chlorinated sodium and potassium facies. Moreover, the groundwater quality in the study area is of poor quality; it is hard and characterized by very high mineralization, The Richards' diagram indicates that the groundwater of the study area are unsuitable for irrigation. The spatial distribution of fluoride ions in groundwater of the terminal complex shows that fluoride levels in Ouargla exceed the World Health Organization standard.

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INTRODUCTION

Water scarcity is one of the major challenges of this century because it is rising as a global issue, where many voices are claiming that water resources might become the most limited resource in the future. There are several factors that cause water scarcity, among them climatic change conditions, population growth, poor irrigation system, and rapid urbanization (Azarnivand and Chitsaz, 2015; Yao et al., 2016; Zhang et al., 2020).

Water scarcity is due to quality issue. Water quality can be degraded by pollution from sewage, agricultural residues, and industrial waste (Yao et al., 2016).

As all North African countries, Algeria is found to be poor in water availability (available less than 500 m^3/person/year)(Jemmali, 2017; Jemmali and Sullivan, 2014).

Furthermore, the water resources vulnerability index reached 87.1% in 2012 characterizing severe water scarcity (Kherbache, 2020).

In the Algerian Sahara, groundwater is the only resource for various uses (drinking, irrigation and industry). This water, contained in the Intercalary Continental (C.I) and the Terminal Complex (C.T) formations, constitutes one of the largest hydraulic reservoirs in the world. Their potential is estimated to 5 billion cubic meters of water (Bouchemal et al., 2011; Youcef and Achour, 2001).

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The quality of groundwater sources are of high risk because of different organic and inorganic pollutant of: (i) various industries, such as: oil and gas production industries, petrochemical industries; (ii) the intensive use of chemical fertilizers in agriculture and the irrational exploitation of water resources, which produce a chemical modification of water, making it unfit for the desired uses (Diamantopoulou and Voudouris, 2008; Haque and Sharif, 2021; Panagopoulos, 2021, 2022). This bad quality is consequently a threat for the environment in general and for human health in particular.

Water supply quality continue to be a major source of human disease and death globally because many of them remain unsafe and vulnerable (Pronczuk and Surdu, 2008; Sobsey, 2006). Nearly 60% of infant mortality is linked to infectious diseases; most of them related water quality and sanitation (Montgomery and Elimelech, 2007).

This research paper aims to understand the present situation of groundwater by evaluating its characteristics taking as case study Ouargla area.

MATERIAL AND METHODS

The state of Ouargla currently comprises 21 Communes and is located at the bottom of the north valley of Oued M'ya, at an altitude of 157 m, corresponding to the geographic coordinates 5° 20' east of longitude and 31° 58' north of latitude(Aissa and Boutoutaou, 2017). Ouargla is characterized by a very arid Saharan climate with moderate winters and a permanent drought where precipitation hardly exceeds 45 mm/year, with average maximum temperatures of 43°C, while the cumulative annual evaporation exceeds almost 60 times that of rainfall (2759 mm/year)(Idir et al., 2014).

The geological formations of the Ouargla region contain two sets of aquifers. The Intercalary Continental, which extends all over the sedimentary basin of the Northern Sahara over an area of 600,000 km^2.The Terminal Complex occupies an area of approximately 350,000 km^2. A third formation, of more modest importance, is added to the previous two. It is the water table (Idder et al., 2014).

The sampling campaign isconcernedthe aquifers exploited (Senonian and Miopliocene) in the eastern region (Ouargla). Sixteen water samples (16) were taken from wells directed to drinking water consumption. These samples were manually taken at the wellhead at each water source.

The groundwater samples were taken in particularly clean plastic bottles with distilled water and drained before being rinsed three times and filled with the water to be analyzed. Each bottle of 1L capacity was labeled with the well code, aquifer type, and date of sampling. The cap was placed in such a way that there were no air bubbles and that it was not ejected during transport.

All samples were kept in an environment of approximately 4°C between sampling and receiving at the laboratory, (we used large cooler and refrigerants).

The physical parameters PH, electrical conductivity (EC) and temperature were measured in situ using a Multiparameter (multi 350 i).

In the laboratory of the centre of Scientific and Technical Research in Physico-chemical Analysis "CRAPC" at the University of Ouargla, the main elements (cations and anions) were analysed according to standard methods described by Rodier et al.(2009).

The total hardness of the groundwater samples was determined by the titrimetric method using eriochrome T black as an indicator; the titration was handled by complexometry of the calcium and magnesium ions with an aqueous solution of disodium salt of ethylene diaminetetracetic acid (EDTA) at a pH of 10.

The Titration of calcium ions was being performed with an aqueous solution of EDTA at a pH between 12 and 13. The indicator used is Murexide, We can reach the concentration of magnesium salt from the following formula $TH=Ca^{+2}+Mg^{+2}$.

The method of titration with sulfuric acid in the presence of methyl orange was used for the determination of the complete alkalimetric title (TAC), which allows to determine the concentration of bicarbonates where at a pH lower than 8.3 the TA is null.

Chlorides were determined by the Mohr method; it is a titrimetric method for the determination of chlorides dissolved in water by a titrated solution of silver nitrate in the presence of potassium chromate.

Sulfates were measured bya UV/Visible Spectrophotometer, the absorbance reading at wavelength λ = 420 nm for a suspension obtained by reaction of sulfates with barium chloride.

The spectrometric determination of nitrates was performed by the sodium salicylate method. Analysis of potassium and sodium using flame emission photometry (JENWAY PFP7), As for fluoride, it was determined by UV-VIS DR 6000 spectrophotometry at λ =580 nm using the reagent SPADNS

The ionic balance is expressed as a percentage of error and is calculated as the relation between the difference of anions and cations and their sum, and 100 multiply the whole:

$$I.B(\%)=[\sum(\text{cations})-\sum(\text{anions})]/[\sum(\text{cations})+\sum(\text{anions})]\times 100 \quad (1)$$

The sum of cations and anions is expressed in equivalence units (either in mEq/L or in °F). The following equation used to covert of mg per litre to mEq per litre, the following equation:

$$mEq/l=(\text{mg per litre}\times \text{valence})/(\text{formula weight}) \quad (2)$$

The value of the percentage obtained can be positive or negative depending on if the greater sum of the ions is that of the cations or the anions. The analysis is considered acceptable with an error of less than 5%. In the opposite case, however, it is rejected or questioned (Walter, 2010).

The spatial distribution of fluoride ions in the groundwater of the terminal complex of the study area was created using geographic information system software (GIS).

The software “DIAGRAMMES” of the Laboratory of Hydrogeology of Avignon allows to draw Piper, stabeler and Richards diagrams, which helps in the presentation of the hydrochemical facies of the studied waters.

RESULTS AND DISCUSSION

Figure 1 shows that all ionic balance values of the chemical analysis results of the groundwater samples are confined between -5 and 5%, indicated that the analysis that has been performed is acceptable (Figure 1).

The results of physical and chemical analyses of groundwater in the study area are presented in Table 1.

Temperature is an important ecological factor for the environment. It is important to know the temperature of the water with a good precision, because it has a role in the solubility of the salts, in the dissociation of the dissolved salts, in the determination of the pH, and for the identification of the origin of the water (Bouchemal, 2017),As can be noticed in Table 1.

The temperatures measured during the month of January vary slightly between 18°C and 23.4°C.This result shows that the temperatures measured on the waters of the terminal complex aquifer have moderately high values, given the sampling period.

The pH of water represents its acidity or alkalinity; at pH 7, water is considered neutral, at pH less than 7, water is acidic, and at pH greater than 7, water is basic. The pH of the water samples varies between 6.94 and 7.46. It is apparent that the pH of the water is neutral with an alkaline character. The values obtained show that the pH of the groundwater of Ouargla is within the standards of potability (6.5-9) (Rodier et al., 2009).

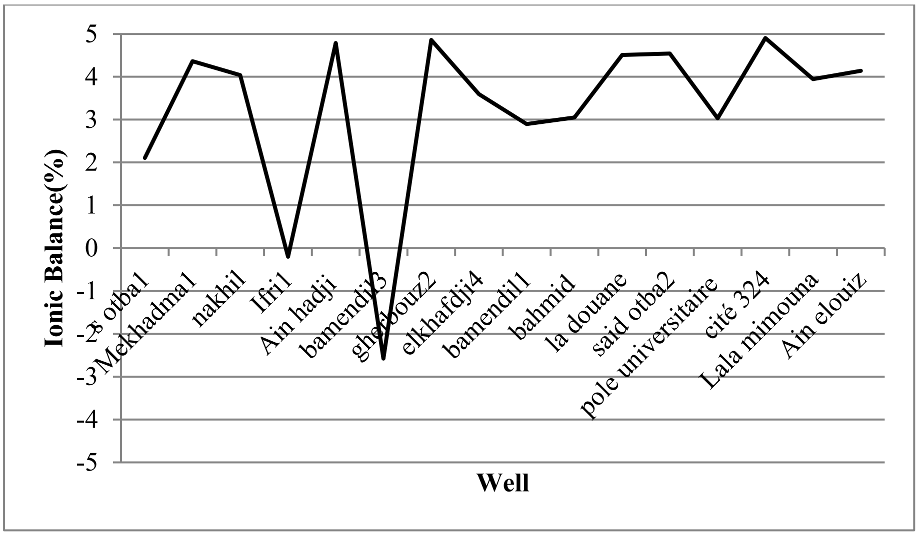

Fig. 1. Ionic balance values of the groundwater samples.

Table 1. The results of the physical parameters of the analysed groundwater samples.

well	aquifer	Easting	northing	T(°C)	PH	EC (µs/cm)
Lala mimouna	Senonian	5,322248	31,953106	18	7,25	2710
gherbouz 2	Senonian	5,314497	31,959255	19,8	7,32	2260
Ain hadji	Senonian	5,318874	31,961021	20,4	7,3	2320
nakhil	Senonian	5,331942	31,955505	21	6,94	2680
bahmid	Miopliocene	5,328932	31,961921	21,6	7,38	3270
Ifri1	Senonian	5,339295	31,964481	22,8	7,34	2680
la douane	Senonian	5,348006	31,957823	23	7,35	3990
Ain elouiz	Senonian	5,349467	31,951366	23,2	7,44	4640
pole universitaire	Senonian	5,292044	31,939044	19	7,46	2600
Mekhadma1	Senonian	5,313031	31,942643	20	7,29	4370
cité 324	Senonian	5,317436	31,948657	20,7	7,13	4240
s otba1	Senonian	5,329535	31,977059	21,2	7,21	2130
said otba 2	Senonian	5,345304	31,977671	21,8	7,32	2480
bamendil 1	Miopliocene	5,284452	31,984003	22	7,3	2150
bamendil 3	Senonian	5,278595	31,983631	22,4	7,35	2310
elkhafdji 4	Senonian	5,258447	31,954676	23,4	7,21	3570

The measurement of electrical conductivity enables evaluating the mineralization of water. As shown in Table 1, the electrical conductivity values of the analyzed groundwater vary between 2130 µS/cm and 4640 µS/cm, according to Rodier et al.(2009). All the measured values of conductivity are above 1000 µs/cm. They indicate excessive mineralization.

Table 2 shows the results of chemical parameters of groundwater in the region of Ouargla for the exploited aquifers.

The total hardness of water is produced by the calcium and magnesium salts it contains. The hardness results mainly from the contact of groundwater with rock formations (Belghiti et al., 2013). The total hardness of the waters samples ranges from 73.4 to 132 °F. This shows that the

Table 2. The results of the chemical parameters of the groundwater samples.

well	TH (°F)	Ca^{+2} (mg/l)	Mg^{+2} (mg/l)	K^{+} (mg/l)	Na^{+} (mg/l)	SO_4^{-2} (mg/l)	Cl^{-} (mg/l)	HCO_3^{-} (mg/l)	NO^{-}_3 (mg/l)
Lala mimouna	88	194	94,8	39,66	789,50	1167,99	1176,21	200,45	21,13
gherbouz 2	78,4	170	86,16	33,30	749,95	1142,33	1058,58	155,92	19,29
Ain hadji	77,2	169	83,88	34,36	749,95	1124,66	1025,65	215,5	19,54
nakhil	92	214	92,4	41,25	710,39	926,61	1227,96	110	24,13
bahmid	114,8	240	131,52	59,02	552,16	1118,63	1096,22	160,8	27,47
Ifri1	88	213,8	82,92	39,66	552,16	945,24	1091,52	110	24,86
la douane	114	270	111,6	65,65	710,39	1065,63	1270,30	197,4	29,33
Ain elouiz	120	297,6	109,44	77,58	789,50	1163,16	1411,45	191,3	31,60
Pole universitaire	73,4	205,6	52,8	43,91	512,61	1077,97	752,77	186,42	22,48
Mekhadma1	125,2	285,4	129,24	84,21	1145,52	956,94	2051,30	142,5	29,63
cité 324	132	291,2	142,08	74,93	710,39	1022,66	1387,92	209,6	30,10
s otba1	74,4	181	69,96	30,12	670,83	1108,27	997,42	190,08	13,32
said otba 2	86,4	199	87,96	35,42	789,50	1056,13	1185,62	196,79	20,24
bamendil 1	76,4	180	75,36	31,71	631,28	1133,00	926,85	177,27	16,90
bamendil 3	80,8	198	75,12	38,60	473,05	1084,31	940,96	99,8	17,03
elkhafdji 4	107,6	251,2	107,52	59,02	591,72	1100,74	1100,93	148,6	28,62

waters of the study area are hard.

Calcium is the dominant element in drinking water. Its levels vary essentially according to the nature of the ground traversed (Rodier et al., 2009). Table 2 shows that all the levels of calcium in the analysed water samples exceed the standard of quality fixed at 100 mg/L.

Magnesium is one of the most common elements in nature; it is a significant element of water hardness (Rodier et al., 2009).As illustrated in Table 2, the results obtained are higher than guideline values (50 mg/L).

Potassium is an essential element in the human body &, it occasionally occurs widely in the environment, including all natural waters (World Health Organization [WHO], 2009). The measured potassium levels ranged from 30.11 mg/L to 84.21 mg/L. Consequently, these values exceed the WHO standard (50 mg/L).

The sodium concentration limit in water that is detectable by tasting depends on the associated anion and the temperature of the solution. At room temperature, the average detection range for sodium is approximately 200 mg/L (L'organisation Mondiale de la Santé [OMS], 2017). The sodium levels in the study area's groundwater range from 473.04 to 1145.51 mg/L, which does not meet the standards.

Sulfate ions (SO_4^{2-}) are related to the major cations: Ca^{+2}, Mg^{2+}, Na^{+}. Most Sulfates of Pb^{2+}, Ba^{2+}, Sr^{2+}, most sulfates are soluble in water (Guerraiche, 2017). As illustrated in Table 2, the sulfate values varied from 926.61 to 1167.98 mg/L, which is not consistent with the WHO standard (250 mg/L).

According to Bashir et al. (2012), residents complained of gastrointestinal tract problems such as diarrhea, nausea, inflammatory bowel disease, and more in areas where consistently higher than guideline values were observed. The chloride values of the analyzed groundwater are very high compared to the guideline value.

The relative values of the alkalimetric title (TA) and the complete alkalimetric title (TAC) make it possible to know the quantities of alkaline or alkaline earth hydroxides, carbonates, or hydrogen carbonates present in water. The alkalinity of water corresponds to the presence of bases and salts of low acids. In natural waters, alkalinity is mostly a consequence of the presence of hydrogen carbonates, carbonates and hydroxides, where waters with a pH lower than 8.3 have a TA that is zero (Rodier et al., 2009).The results obtained indicate that all the bicarbonate values [HCO_3-] of the water samples exceed the WHO standard.

The main risk to human health associated with the ingestion of nitrates is related to the ability of the human body to transform nitrates to nitrites. This reduction of NO_3^- to NO_2^- , is done by bacteria with nitrate reductase activity (Banas&Lata ,2006).The nitrate concentrations of the groundwater samples vary between 13.31 and 31.60 mg/L. See Table 2. It is clear that the results obtained are in accordance with the WHO standard.

Figure 2 indicate that the groundwater of the terminal complex of the study area is characterized by a predominance of chloride ions over bicarbonate and sulfate ions. As for the cations, sodium is the dominant cation, followed by calcium.

The Piper diagram of the major elements of the waters of the terminal complex (Ouargla) reveals that the waters of this aquifer present a sodium chloride and potassium facies. Therefore, the dominant ions are chlorides for the anions Cl^->$SO4^{-2}$>HCO_3^- and sodium and potassium for the cations Na^+ and K^+> Ca^{+2}> Mg^{+2}(Figure 3).

Excessive consumption of fluoride can cause (skeletal fluorosis, dental fluorosis). Skeletal fluorosis is a bone and joint condition. Enamel fluorosis is a dose-related mottling of enamel that can range from mild discoloration of the tooth surface to severe staining and pitting (National Research Council [NRC], 2006).

According to a study in the Algerian northern Sahara, the adverse effects of long-term human exposure to fluoride are manifested by the appearance of dental fluorosis.

The spatial distribution map of fluoride ions (Figure 4) reveals that the groundwater of the Ouargla terminal complex contains fluoride concentrations varying between 1.11mg/L and

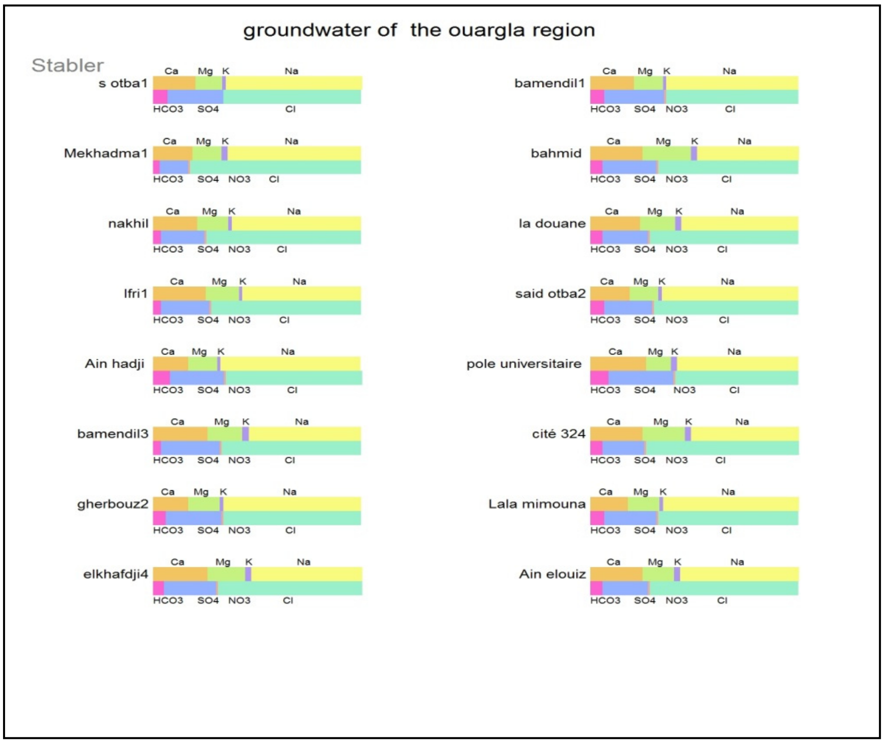

Fig. 2. Ion classification of groundwater samples using Stabler diagram.

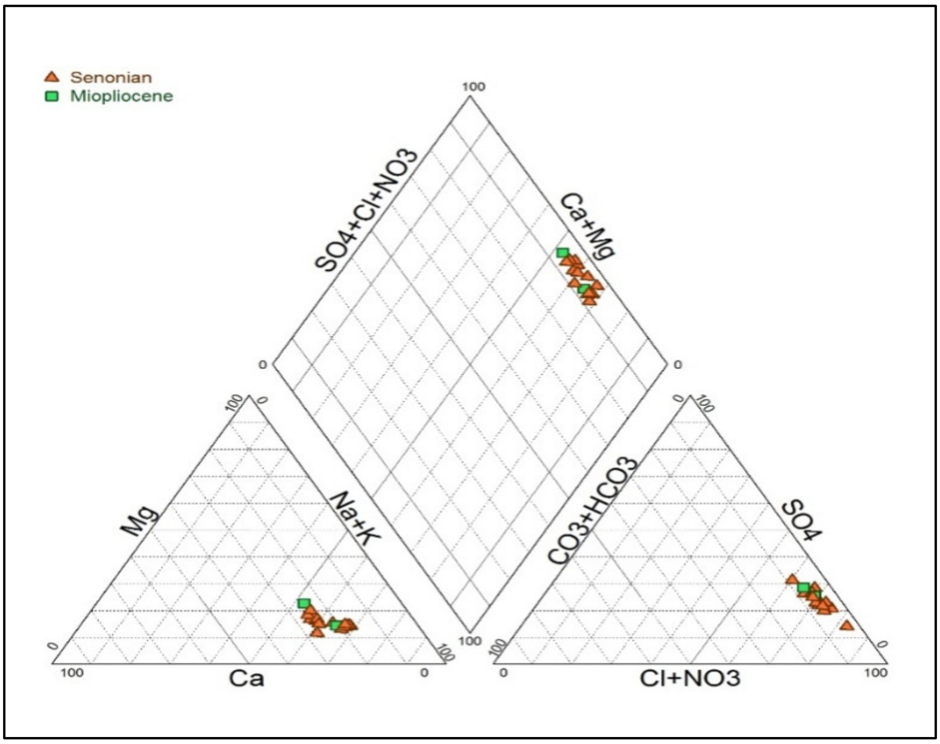

Fig. 3. Piper diagram of the groundwater of the study area.

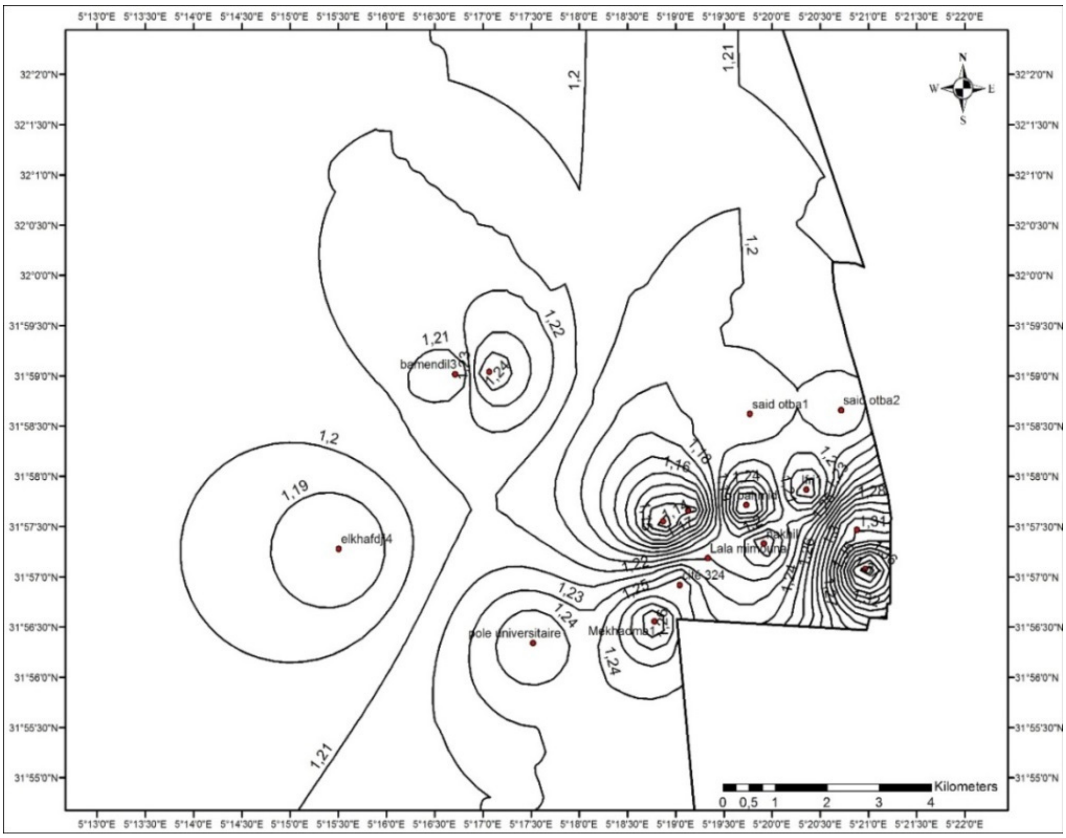

Fig. 4. The spatial distribution of fluoride ions in the groundwater of the terminal complex of the study area

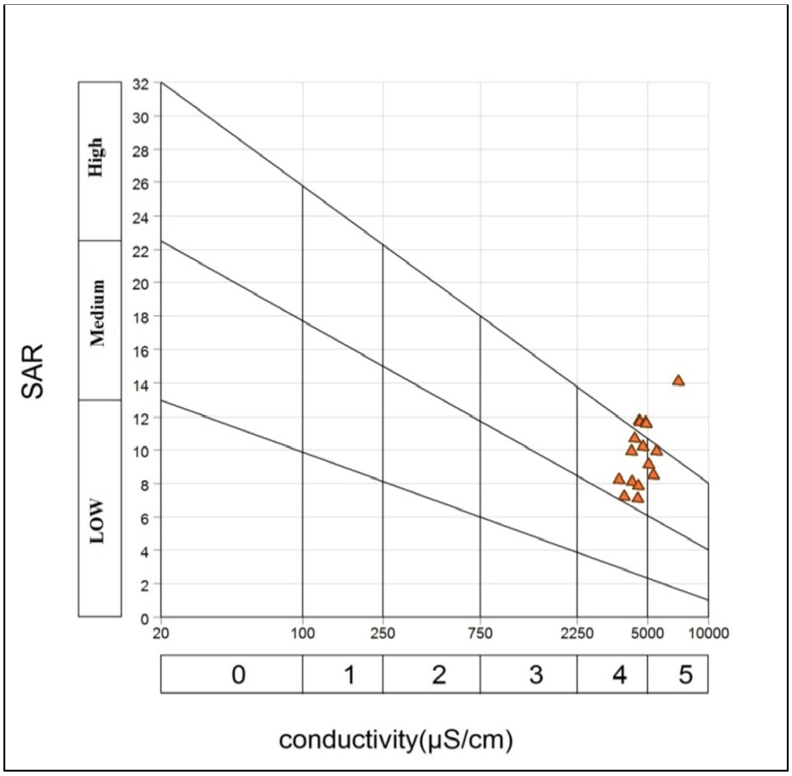

Fig. 5. Richards' diagram of the terminal complex groundwater in the study area.

1.39mg/L. These values are contrary to the standards of the WHO, which state that in a hot climate, Recommended control limits range from 0.6 - 0.8 mg/L for temperatures of 26.3-32.6° C to 0.9-1.7 mg/L for temperatures of 10-12° C (WHO, 2008).

The suitability of the groundwater for irrigation has been evaluated based on the sodium adsorption ratio (SAR).

Sodiumconcentrationisanimportantcriterioninirrigation-waterclassificationbecausesodium reacts with the soil to create sodium hazards byreplacing other cations. The extent of this replacement is estimated by Sodium Adsorption Ratio (SAR) (Goswamee et al.,2015).

The sodium adsorption ratio of water gives the measure of suitability of water for irrigation with respect to sodium (alkali) hazard (Adimalla et al., 2018; Goswamee et al.,2015; Nayak et al., 2022; Vasanthavigar et al.,2010). It is given by the formula

$$SAR= [Na^{+}]/ \{([Ca^{+2}] + [Mg^{+2}])/2)\}^{1/2} \quad (3)$$

Where

SAR = sodium adsorption ratio (millimole/litre) 1/2

Ca^{2+}= -calcium ion concentration, me/l

Mg^{2+} = magnesium ion concentration, me/l

NOTE: me/l = mill equivalent/litre

According to the Richards diagram shown in Figure 5, the groundwater samples are mainly located in the C4S3 (high salinity with high sodium), and C5S3 (very high salinity with high sodium) class except for 2 samples that are located in the C4S4 (high salinity with very high sodium) class and one sample is located in the C5S4 (very high salinity with very high sodium) class.

The results obtained after the interpretation of the Richards diagram showed that the groundwater of the terminal complex of Ouargla is unsuitable for irrigation because of a very high salinity and the hazard of sodium.

CONCLUSION

We have studied the physicochemical quality of groundwater in the region of Ouargla. This investigation has concerned the exploited water table (Terminal Complex). To carry out the determination of the physico-chemical parameters, we have used different analytical methods. The waters analyzed in this study have an ionic balance lower than 5%, thus confirming the reliability of the chemical analysis. Obtained results show that the groundwater exploited in the Ouargla region is of poor quality; it is hard and characterized by very high mineralization. Moreover, the fluoride levels exceed the WHO standard. The groundwater of the Ouargla region presents a sodium and potassium chloride facies. Furthermore the dominant ions are chlorides for the anions Cl^{-}>$SO4^{-2}$>$HCO3^{-}$ and sodium and potassium for the cations Na^{+} and K^{+}> Ca^{+2}> Mg^{+2}. In addition, the results obtained indicate that the groundwater in the study area is not suitable for irrigation.

The results of this study giving a visionary anticipatory for groundwater pollution hazards, the researchers strongly recommend of intensify efforts to protect the groundwater quality and sustainability to protect human health and the ecosystem.

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The present research did not receive any financial support.

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication &/ or falsification, double publication &/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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