

GROUNDWATER CONTAMINATION BY NUTRIENTS: CASE OF GADAÏNE-AIN YAGHOUT PLAIN (NORTH OF BATNA), ALGERIA.

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Abstract

In Algeria, groundwater face a risk of degradation due to uncontrolled releases, intensive and indiscriminate use of chemical fertilizers in agriculture as well as disorderly exploitation of this resource. The consequences are a chemical modification of the water, making it unsuitable for desired uses, which makes the field of water increasingly vital and fragile requiring for its preservation monitoring and control of the management of this resource. To determine the causes of the progressive degradation of the chemical quality of groundwater, this study was devoted to the knowledge of the geological and hydrogeological characteristics of the plain Gadaïne - Ain Yaghout (North of the city of Batna) and interpretation of physico-chemical analysis of water samples with highlighting the various sources of pollution and their impact on groundwater of the plain.

Keywords: Chemical fertilizer, physico-chemical analyzes, pollution, groundwater, Algeria.

Résumé

En Algérie, les eaux souterraines sont confrontées à un risque de dégradation, à cause des rejets non contrôlés, de l'utilisation intensive et anarchique des engrais chimiques dans l'agriculture ainsi qu'une exploitation désordonnée de cette ressource. Les conséquences sont une modification chimique de l'eau, qui la rend impropre aux usages souhaités, ce qui rend le domaine de l'eau de plus en plus vital et fragile, nécessitant pour sa préservation le contrôle et la maîtrise de la gestion de cette ressource. Afin de déterminer les causes de cette dégradation progressive de la qualité chimique des eaux souterraines, nous avons consacré cette étude à la connaissance des caractéristiques géologiques et hydrogéologiques de la plaine de Gadaïne – Ain Yaghout (Nord de la ville de Batna) et l'interprétation des analyses physico-chimiques d'échantillons d'eau avec une mise en relief des différentes sources de pollution et leur impact sur les eaux souterraines de cette plaine.

Mots clés: Engrais chimiques, analyses physico-chimiques, pollution, eaux souterraines.

المخلص

في الجزائر، المياه الجوفية تواجه خطر تدهور نوعيتها بسبب مياه الصرف غير المراقبة والاستخدام المكثف والعشوائي للأسمدة الكيماوية في الزراعة، فضلا عن الاستغلال غير المنظم لهذا المورد. ينجرعن ذلك التعديل الكيميائي للمياه، مما يجعلها غير صالحة للاستخدامات المطلوبة، بالإضافة إلى سيادة الجفاف في بعض المناطق مما جعل مجال المياه حساس وهش على نحو متزايد متطلبا بذلك للحفاظ عليه المراقبة والتحكم في إدارة هذا المورد. بهدف تحديد الأسباب التي أدت إلى التدهور التدريجي للنوعية الكيميائية للمياه الجوفية، فقد خصصنا هذه الدراسة إلى معرفة الخصائص الجيولوجية والهيدروجيولوجية لسهل غداين – عين ياغوت (شمال مدينة باتنة) و تفسير التحليلات الفيزيوكيميائية لعينات المياه مع إبراز مختلف مصادر التلوث وتأثيرها على المياه الجوفية في هذا السهل.

الكلمات المفتاحية: الأسمدة الكيماوية، التحليلات الفيزيوكيميائية، التلوث، المياه الجوفية.

INTRODUCTION :

Since the 1950s, nitrate (NO_3) contamination of groundwater has become a significant environmental problem in Algeria. Agriculture can have a significant impact on the quality of groundwater. Nitrate pollution of shallow aquifer is of high concern as it may have negative impacts on water supply and ecosystems [1, 2]. Nitrate (NO_3) is a familiar pollutant in groundwater. Large amounts of nitrate in drinking water are a cause of methemoglobinemia (also known as blue baby syndrome), a blood disorder primarily affecting infants under 6 months of age [3, 4].

Groundwater generally represents an excellent source of provision of drinking water. Natural filter constituted by the geological materials often produces high quality water. However, maintaining this relative advantage requires that measures be taken to preserve sustainably the quality of the natural reservoir of provision. The proposed study is set in a semi-arid region of Algeria, in the southern of constantinian high plains. This region is the plain of Gadaïne - Ain Yaghout located in the North of the city of Batna (Fig.1). In this study, hydrogeologic data from the aquifer system will be used to determine the main factors and mechanisms controlling the chemistry of groundwater in the study area (NE Algeria) and to identify the origins of water degradation.

STUDY AREA:

Gadaïne - Ain Yaghout plain is located in northern Africa between longitudes $35^{\circ}42'$ - $35^{\circ}54'$ North and latitudes $6^{\circ}15'$ - $6^{\circ}30'$ East, at an average elevation of 820 m above sea level. It is a part of southern fringe of the constantinian high plains (Algeria). It is situated between the constantinian neritic in the North and the Atlasic Mountains (Aurès) in the south. The total area of the plain is about 416 km² and more than 8981 inhabitants are living in this area [5]. "Chott" is a North African term for a salt flat within a hydrologically closed basin [6], and the elevation of a Chott surface is controlled by the position of the water table and associated capillary fringe and also by the climate impact (water cycle, hydrology and hydrogeology). The drainage network is not very dense, it is composed of the El Madher and Ben Zerhaïb non-perennial streams known locally as "wadis". These subdendritic wadis collect surface runoff from the surrounding mounts of the study area. The surface water of these wadis is carried to the endorheic depression of Chott Gadaïne in the South-West and to Chott Tinsilt in the North-East part of the study area.

CLIMATE:

The study area has a Mediterranean climate, with hot summers months (June, July and August) and cold winters months (December, January and February). It shows a mean annual rainfall of about 334 mm/year (data

based on observations from 1984 to 2000) with a maximum amount of rainfall from March to May. The maximum rainfall amounts are associated with the highest elevations of the study area. The potential evapotranspiration is about 785 mm/year [7]. The annual average temperature is around 14°C, with a maximum from June to August (32°C) and a minimum between December and February (0.2°C) [7].

GEOLOGICAL SETTING:

The study area belongs to the external domain of the Maghrebide chain of which we find (Fig. 1):

- In the North, the whole south Sétifian allochthon composed by the scales of Guedmane mount, Tizourit mount and Merzeguène mount which have a Cretaceous age [8] ;
- In the South and the East, the reliefs belong to north aurésian parautochthone and autochthone formations constituted by scales of AinYaghout mountains (Tarbennt, Haouia and Azem mounts), Tafraout, Koudiat Tfouda and Sarif mounts which have a Jurassic - Cretaceous age. These mountains consist mainly of rocky formations with carbonate dominance and forming a continuous lithological series from Lias to Cretaceous [8] ;
- A mio-plio-quadernary cover occupies the entire plain, formed by lacustrine limestone, clays, conglomerates, alluvium and polygenic glacia in particular and salty soils bordering chotts [8].
- The Triassic formations outcrop mainly in the East of the plain, made of colorful marl, breccia gypsum and dolomite limestone [8].

HYDROGEOLOGY:

Determining of the different aquifers in the plain of Gadaïne - Ain Yaghout was carried on the lithostratigraphic and structural analysis of geological formations as well as geophysical data. From this analysis, hydrogeological cache of the plain reveals two aquifers [7]:

- The Mio-plio-quadernary aquifer is a heterogeneous aquifer constituted essentially by lacustrine limestones, conglomerates and alluvium. These formations occupy almost all the study area except reliefs;
- The deep aquifer of Jurassic-Cretaceous age in which its carbonate formations outcrop in the border of the plain forming massifs.

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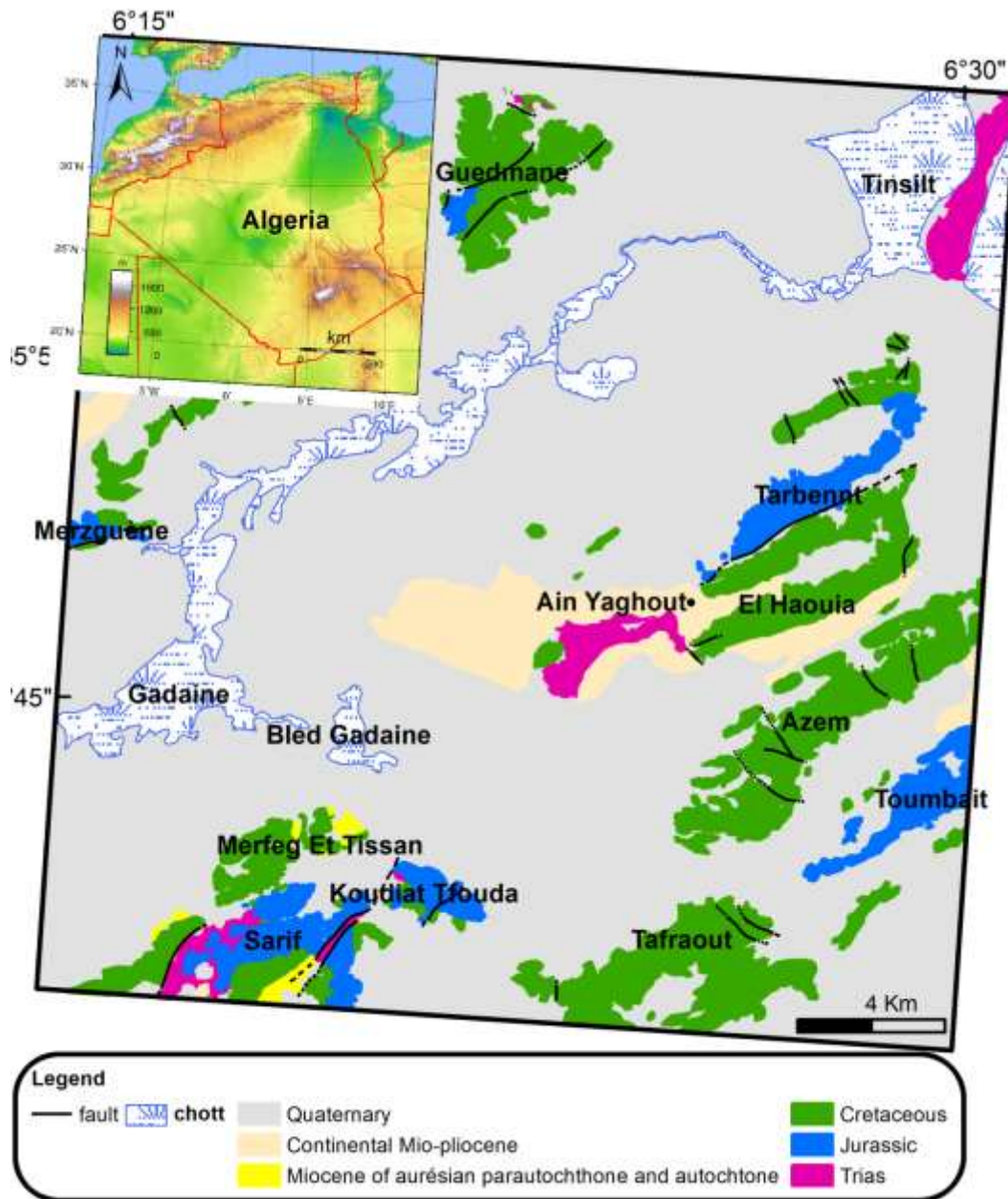


Figure 1: Location and geological map of the study area. (J. M. Vila, 1977)

Our study area contains 29 wells and 16 boreholes dispersed in the plain. They are destined for drinking water supply, irrigation and other domestic uses. The water points permit us to realize the piezometric map of May 2009 (Fig. 2) which shows that the flows are oriented southeast to the northwest in the east of the plain, and from the Northwest to the southeast in the west. These flows converge to the drainage area and thereafter to Tinsilt chott.

MATERIALS AND METHODS:

Sampling and analysis

In Mai 2009, a total of 25 groundwater samples have been obtained from active boreholes (13) and wells (12) capturing the mio-plio-quaternary aquifer. Immediately after sampling, temperature, pH and electrical conductivity (EC) were measured in the field using a multi-parameter (Model: HANA, Hi 991.301) to ensure the exclusion of the atmosphere effect, contamination and to improve the stability of the measurement.

Subsequently and in the same period (Mai 2009), the samples were analyzed in the laboratory for their chemical constituents such as calcium, magnesium, sodium, potassium, chloride, bicarbonate, sulfate, nitrate, nitrite and ammonium. For this chemical analysis the samples have been collected in plastic bottles. This was achieved using standard methods as suggested by the American Public Health Association [9, 10]. Ca^{2+} , Mg^{2+} , HCO_3^- , NH_4^+ and Cl^- were analyzed by volumetric titrations. Concentrations of Ca^{2+} and Mg^{2+} were estimated titrimetrically using 0.05 N EDTA and 0.01 N and those of (HCO_3^- , NH_4^+) and Cl^- by H_2SO_4 and $AgNO_3$ titration, respectively. Concentrations of Na^+ and K^+ were measured using a flame spectrophotometer (Model: PYE UNICAM, Philips) and spectrophotometer (Model: HACH, DR 2000, wave 766.5), respectively, and that of sulfate (SO_4^{2-}) by using colorimeter (JENWAY 6051, Csuflate). Nitrate (NO_3^-) and nitrite (NO_2^-) was analyzed by using spectrophotometer (HACH, DR 2000). Standard solutions for the above analysis were prepared from the respective salts of analytical reagents grades. The accuracy of the chemical analysis was verified by calculating ion-balance errors where the errors were generally less than 5%.

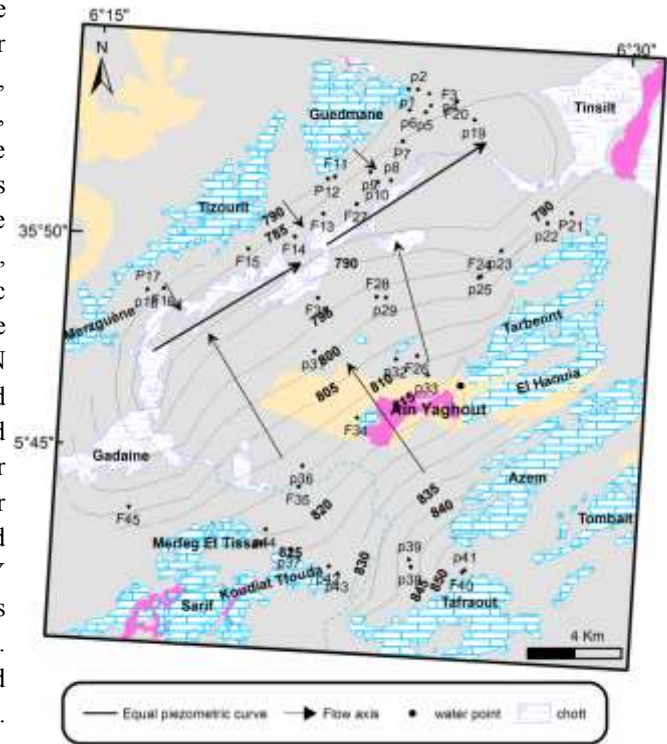


Figure 2: Piezometric map of the plain of Gadaine - Ain Yaghout, (Mai 2009). (Dib, 2009)

RESULTS AND INTERPRETATION:

Table 1 presents the statistical summary of all the parameters analyzed. The mean concentrations of the major ions in the Mio-Plio-Quaternary aquifer are within the WHO (2008) guidelines for drinking water [11].

Table 1 : Chemical summary of shallow groundwater in the study area, (Mai 2009).

	EC	T	pH	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	NH ₄ ⁺	Cl ⁻	SO ₄ ²⁻	HCO ₃ ⁻	NO ₃ ⁻	NO ₂ ⁻
WHO	750	25	6.5-9.5	100	50	20	100	0.5	250	250	300	50	0.1
Mean	4160.84	19.42	7.13	184.37	135.42	489	5.68	0.21	937.61	348.7	400.8	24.60	0.04
Min	1941	14	6.74	79	69	65	1	0.00	226	93	186	0.62	0.00
Max	7260	24	7.90	279	264.38	1120	11	0.95	1913	591	1860	112.92	0.29
SD	1338.23	2.85	0.33	54.76	51.67	287.63	2.66	0.31	440.91	111.3	319.7	28.38	0.08
Cv	0.32	0.15	0.05	0.30	0.38	0.59	0.47	1.52	0.47	0.32	0.80	1.15	1.84

All values are in mg/l, except pH, T (°C) and EC (µ.Siemens/cm) WHO (2008).

Physical Parameters of Water samples:

Temperature of groundwater samples ranges from 14°C to 24°C. These temperatures do not exceed the WHO standard. This thermal variation may be function of the depth of the aquifer which is little or not deep this implies that the water temperature will be influenced mainly by climatic variations. The measured pH ranges between

6.74 and 7.90 with a mean value of 7.13. This shows that the groundwater of the study area is mainly of neutralin nature. The comparison of measured pH with the equilibrium pH shows that the majority of water points are characterized by encrusting water.

The electrical conductivity of groundwater samples ranges from 1941 to 7260 µS/cm therefore remain above drinking water WHO standards. The conductivity

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increases generally in the direction of groundwater flow in the east and the south of the plain (Fig.3). The salinity increases in the direction of groundwater flow, this is due to leaching of traversed formations (especially saliferous Triassic) and evaporation (when the piezometric level is close to the soil surface).

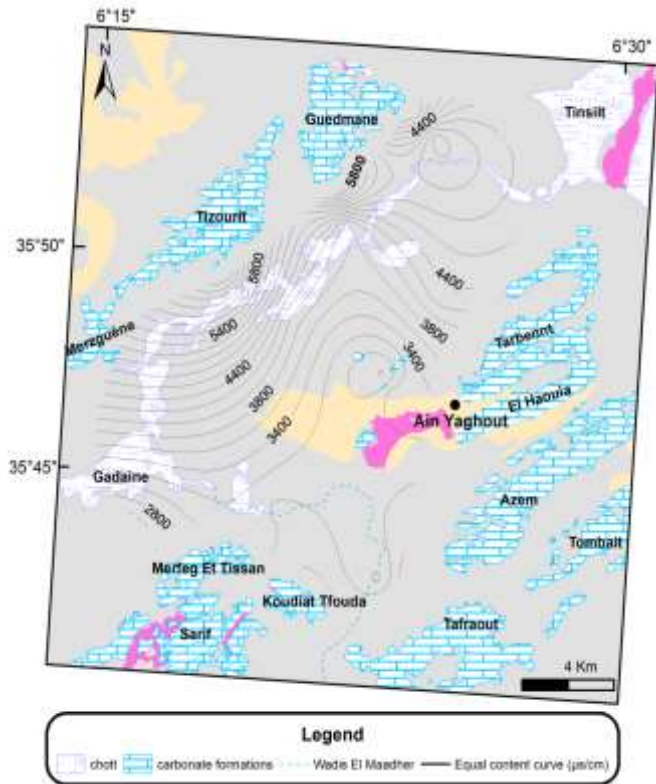


Figure 3: Conductivity map of the plain of Gadaïne - Ain Yaghout. (Dib, 2009)

Near a small Triassic outcrop in the village of Ain yaghout, the electrical conductivity is less than 3200 $\mu\text{S} / \text{cm}$, this value increases to the area between the chotts and western reliefs of the plain. In this area, the groundwater is influenced in largely by the saliferous formations constituting the chotts than the carbonate formations of these reliefs.

General hydrochemistry:

The rock dominance of the major-ion chemistry in the region provides an insight of chemical weathering in the aquifer, since weathering of different parent rocks (e.g. carbonates and evaporites) yields different combinations of dissolved cations and anions to solution. For example, HCO_3^- is from carbonates, SO_4^{2-} and Cl^- are from evaporites [12].

In the recharge area, the dominant geochemical process is the dissolution of carbonate minerals, which contributes the Ca^{2+} , Mg^{2+} and HCO_3^- to the groundwater. Chemical analysis of groundwater shows a general decrease of Ca^{2+} , Mg^{2+} , HCO_3^- and pH as the groundwater moves away from the recharge area which could be the result of

carbonate (limestone, dolomite). Localized gypsum dissolution along the flow path contributes both Ca^{2+} and SO_4^{2-} to groundwater. The Ca^{2+} released by the dissolution of gypsum leads to the precipitation of additional calcite and liberates CO_2 , which leads to a slightly low pH and super-saturation or near equilibrium of groundwater with calcite. This phenomenon is referred to as common-ion driven precipitation or common ion effect [13, 14]. The common ion effect of gypsum dissolution and calcite precipitation is often accompanied by dolomite dissolution, leading to the observed increase in Mg^{2+} in groundwater.

The concentrations of SO_4^{2-} , Cl^- , Na^+ and K^+ values increase from the recharge area towards chotts area where saliferous formations (gypsum, clay and marl) are prevalent. Concentrations of Na^+ and Cl^- in groundwater provide evidence that halite dissolution (Triassic formations) is the major process controlling Na^+ and Cl^- in groundwater. It is known that Cl^- is a highly mobile chemical species interacting weakly with the host rock, which is part of the so-called conservative elements, frequently used as tracers for the study of the hydrological cycle.

Sources of major components in the groundwater:

Cl^- and SO_4^{2-} are the major anions and Ca^{2+} and Na^+ are the major cations in shallow groundwater in Gadaïne-Ain Yaghout plain. The relative abundance of the ions is $\text{Na}^+ > \text{Mg}^{2+} > \text{Ca}^{2+} > \text{K}^+ > \text{NH}_4^+$ (on molar basis) and $\text{Cl}^- > \text{HCO}_3^- > \text{SO}_4^{2-} > \text{NO}_3^- > \text{NO}_2^-$. The maximum Ca^{2+} and Mg^{2+} concentrations of 279 mg/l and 264.38 mg/l respectively are, however, higher than their respective WHO standards [11] of 100 mg/l and 50 mg/l. The common source of calcium and magnesium in the groundwater is limestone and dolomite in the sedimentary rocks. Carbon dioxide in atmosphere and soils allows the dissolution of carbonate rocks, this gives bicarbonate ions HCO_3^- in the groundwater. Its maximum concentrations near massives of study area reaches about 1860 mg/l. The mean sodium and potassium concentrations in the groundwater are 489 mg/l and 5.68 mg/l, respectively.

The chloride ion is widely distributed in natural water. Most Na^+ and Cl^- in the groundwater is from three sources including ancient seawater entrapped in sediment, solution of halite and related minerals in evaporate deposits existing in salt chotts and triassic formations in the region. The value of the chloride in the study area ranges between 226 and 1913 mg/l, these values make it the principle anion in the investigated water.

The most extensive and important occurrences of sulfate ions in the investigated water are from the dissolution of sedimentary rocks such as gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and/or anhydrite (CaSO_4). During weathering, the sulfides, which are in contact with water, are oxidized to yield sulfate that is carried off in water. In the study area, the

leaching of sulfate from the upper soils causes the sulfate to be the second principle anion in the groundwater. Further addition of sulfate to the groundwater comes from the leaching of fertilizers used for agriculture. The value of SO_4^{2-} in the study area ranges between 93 and 591 mg/l. Almost 96% of the samples exceeded the desirable limit of Cl (250 mg/l), and 84% of them exceed that of SO_4^{2-} (250 mg/l) [11].

Groundwater facies:

The results of chemical analysis are reported in the Piper diagram [15] that shows us two point clouds (Fig. 4). A first group represents 60% of the points having a "chloride sodium and potassium or sodium sulfate" facies. A second group represents 40% of the points that have "chloride and calcium sulfate and magnesium" facies. Both facies indicate the influence of salt formations mainly Triassic on groundwater salinity in the plain of Gadaïne - Ain Yaghout.

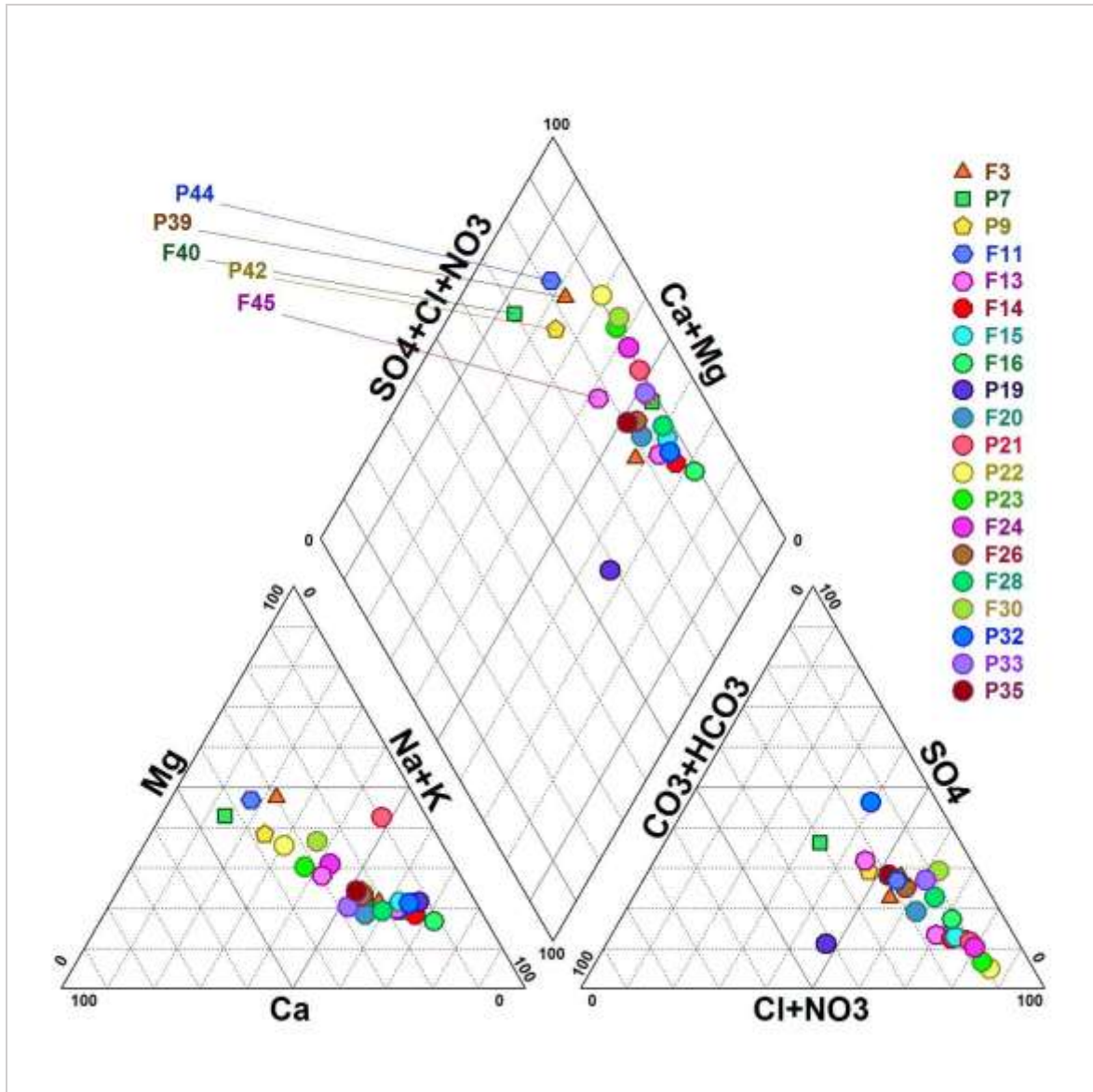


Figure 4: Piper plot depicting the chemical composition of the groundwater samples of the study area, (Mai 2009).

Ionic relations:

To precisely identify the principal origin of Na^+ and Cl^- ions, bivariate plot of Na versus Cl was established (Fig. 8). Most of water samples are grouped along a slightly inclined line ($R = 0.86$) relative to the line of halite dissolution ($R = 1$). That is suggesting that these ions mostly derived from the same origin; likely in relation with the dissolution of halite.

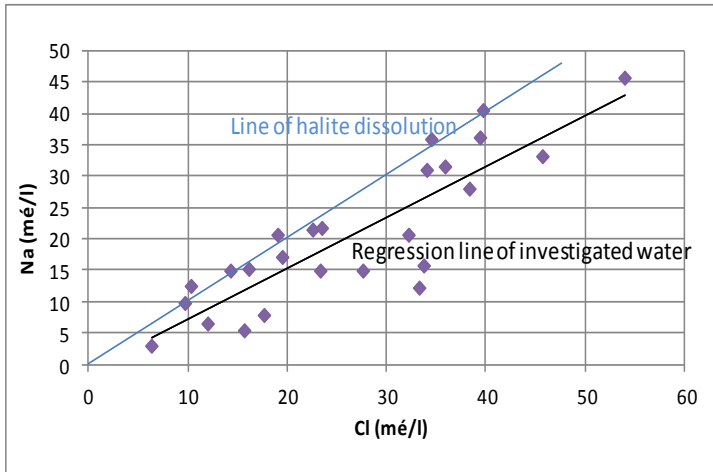


Figure 8: Na^+ vs Cl^- relationship.

In addition, excess chloride is remarkable; this indicates an additional origin which could be the discharge of sewage into septic tanks.

From the graph (Fig. 9), which reflects the " $r(\text{Ca}^{2+} + \text{Mg}^{2+}) / r(\text{SO}_4^{2-} + \text{Cl}^- + \text{Na}^+)$ - electrical conductivity" relationship, we note that the majority of water points congregate in a way to form an evaporite pole where the " SO_4^{2-} , Cl^- and Na^+ " evaporitic ions are dominant for a conductivity greater than $1900 \mu\text{S} / \text{cm}$.

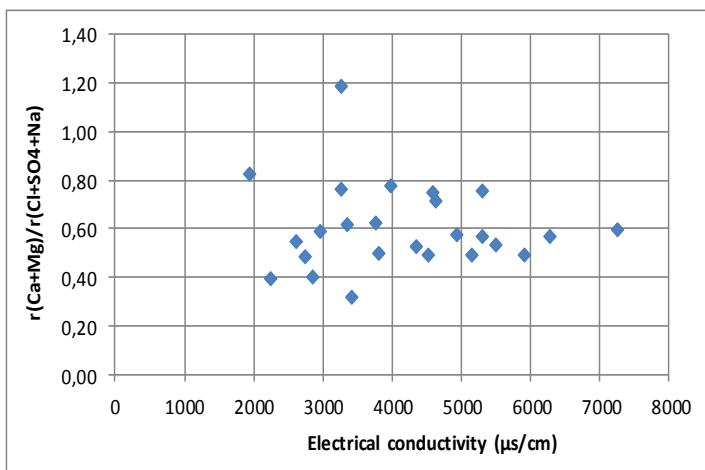


Figure 9: Relationship $r(\text{Ca}+\text{Mg})/r(\text{Cl}+\text{SO}_4+\text{Na})$ vs electrical conductivity.

Spatio-temporal distribution of nutrients:

For the maps representing the spatio-temporal distribution of nitrates the Krigging geo statistical interpolation method was used. Universal Krigging is considered the best linear unbiased estimation for spatial interpolation [16]. The maximum NO_3^- concentrations in May and September are 112.92 mg/l and 133.09 mg/l respectively, however, are higher than their respective WHO (2008) standards. These concentrations are located in the East (near Tarbennt mount) and the center of the plain (Fig. 5). The oxidation of ammonium and nitrite by bacteria can form nitrates (nitrification phenomenon).

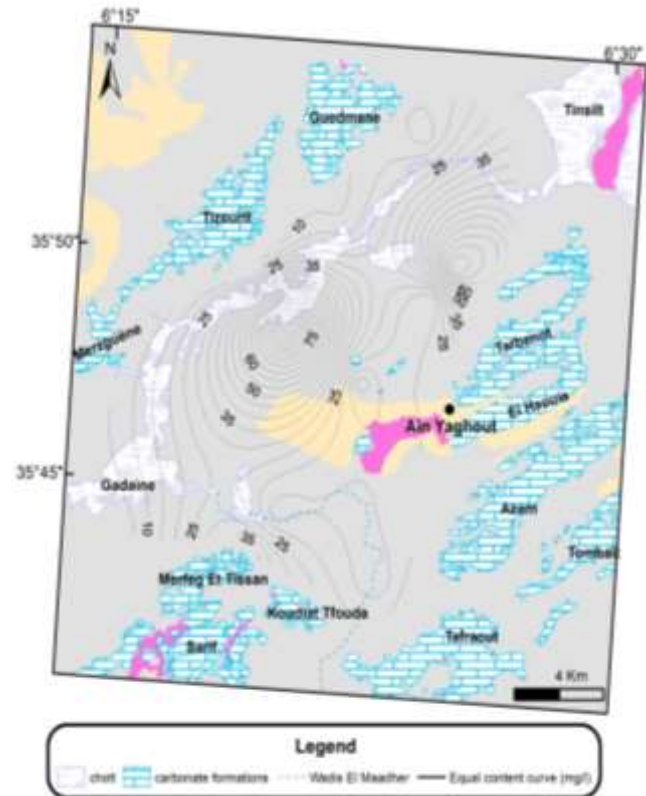


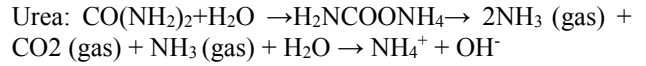
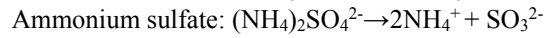
Figure 5: Map of nitrate concentrations (Mai 2009). (Dib, 2009)

The minimum NO_2^- concentrations are 0.29 mg/l and 0.41 mg/l during May and September respectively, in wells located north and east of the study area (Fig. 6). All forms of nitrogen (organic nitrogen, ammoniacal and nitric) can be at the origin of nitrites, either by incomplete oxidation of ammonia, or by reduction of nitrates under the influence of bacterial action. During May, the maximum ammonium concentration is 0.96 mg/l . This concentration increases in September to reach about 1.03 mg/l . These concentrations are higher than the drinking water standards ($0.5 \text{ mg} / \text{l}$) in wells located in Northeast and center of the study area (Fig. 7). The ammonium may come from the reduction in reduced conditions of nitrogen forms (nitrite and nitrate) coming from different sources.

Nutriments origins:

Groundwater does not contain originally nitrogen compound, it is artificially increasing of the quantity of combined nitrogen available in the soil which creates an imbalance between intake and consumption and produces an excess of nitrogen which is entrained in to the groundwater. In our study area, this excess is related to the excessive application of chemical fertilizer of a simple type (containing a single element among three: N or P or K) consisting essentially of nitrogen.

When these fertilizers are applied to the soil, they undergo chemical and biological transformations that end up by releasing into the soil solution (some soil moisture is needed) nitrogen as NO_3^- and/ or NH_4^+ , according to the following reactions [17]:



The ammonitrate and ammonium sulfate produce nitrate and ammonium. Nitrates are immediately absorbed by the plants in case of need, or they can be trained in depth by rainfall or irrigation water because their negative electrical charge does not allow them to be retained by soil colloids. The soil nitrates can also be lost in the form of nitrogen oxides (NO , NO_2 ...) by denitrification, in case of water excess.

The ammonium ions can be either absorbed but to lesser degrees than nitrates by the plants or transformed to nitrate by nitrification, or fixed by the negative electric charges of soil colloids. Under certain conditions, ammonium can turn into ammonia and volatilize.

The urea transformation requires the intervention of an enzyme capable to break the links C-O. This is provided by a wide range of microorganisms present in the soil. The nitrogen is first liberated as ammonia which either to volatilize as a gas or dissolved in the soil water, to give the ammonium which follows the same steps as the ammonium liberated by the sulphate ammonium or the ammonitrate. The application of these fertilizers has the potential to pollute not only the soil but also the groundwater by the loss of nitrates and their migration to these waters [18, 19].

The livestock effluents come in second position in nutrient inputs to agricultural lands. The nutrient content of the effluents depends on the type of farming, pasture systems and nutrient content of different foods and forages used for livestock. Thus, the method of storage of animal waste in which their injection into the soil rather than their spreading on the surface reduces the risk of ammonia volatilization and increases the risk of leaching. Some fungi and bacteria decompose plant and animal waste and then produce ammonia which will dissolve to form ammonium. In addition, domestic sewage are

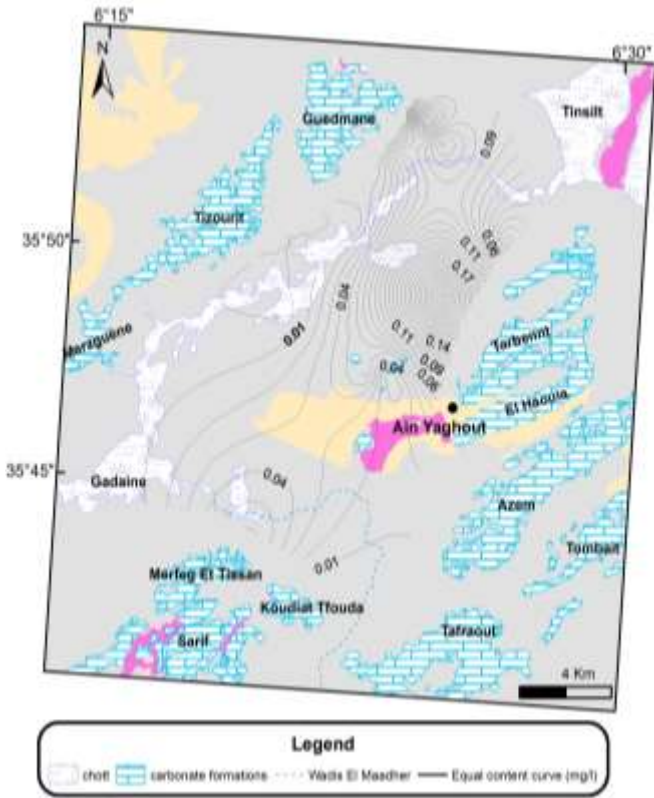


Figure 6: Map of nitrites concentrations (Mai 2009). (Dib, 2009)

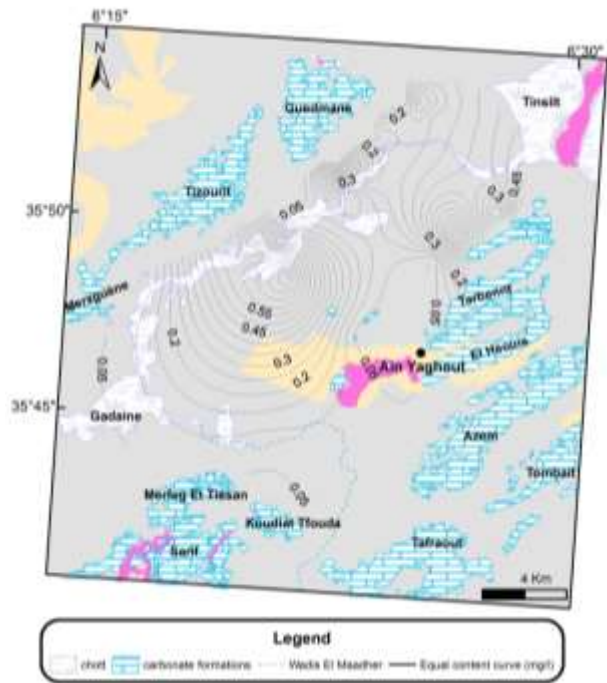


Figure 7: Map of ammonium concentrations (Mai 2009). (Dib, 2009)

discharged directly either in septic tanks (not conforming to standards) or in El Madher wadi and other secondary wadis (without any treatment). These waters contain urine consists mainly of nitrate and ammonium (which is slowly oxidized to nitrate using nitrifying bacteria). Thus, anarchical releases (without any site study) of household waste can cause many pollution and dangers by impairing the quality of groundwater by infiltration, the air quality and aesthetic of the environment.

CONCLUSION

For some samples analyzed, the concentrations of major chemical elements (Na^+ , Mg^{2+} , Ca^{2+} , Cl^- , HCO_3^- and SO_4^{2-}) and nutrients, have exceeded the drinking water standards set by WHO (2008). These samples were mainly taken near agriculture areas.

The concentrations of NO_3^- , NO_2^- and NH_4^+ during September are higher than those of May. Thus, the risk of groundwater nutrients contamination is on increasing.

During the observation period, water quality degradation was observed. It is due mainly to the following factors:

- Geology: the lithological description showed heterogeneity in geologic formations (clays, lacustrine limestones, conglomerates, alluvia and evaporites rich in Cl^- , Na^+ , SO_4^{2-} and Ca^{2+}) component the superficial aquifer of the region which favors the groundwater mineralization.
- Climatic factors: the semi-arid regions are generally characterized by a wet winter and a hot and dry summer with high temperatures induces high evapotranspiration and consequently an increase in the salinity which may explain the degradation of waters quality.
- Anthropic factors: poor water quality can be explained by discharges of wastewater in wadis and septic tanks, the use of nitrogen fertilizers and manure or animal wastes.

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