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Origin of water salinity in Annaba aquifer system, North-Eastern Algeria

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Abstract

The Annaba area hosts in its underground a water potential of great importance. In fact, it is one of the Algerian plains where groundwater is subject to over-pumping. Moreover, the expansion of farmlands and the development of the neighboring agglomerations required a massive pumping of water, thereby leading to the change in hydrodynamic regime of groundwater and to the degradation of its quality. On the basis of boreholes and physicochemical data, the three major factors responsible for the evolution of chemical quality of water observed at the aquifer were identified: (1) mineralization due to natural and anthropogenic processes (responsible for the increase in the contents of chlorides, sodium, calcium and magnesium), (2) the oxydoreduction conditions due to the passage of the water table from an unconfined aquifer or even semi-confined to a confined aquifer (responsible for the reduction of nitrates and (3) pollution of groundwater by nitrates in areas where the water table is shallow and in the absence of a protective clayey cover.

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Introduction

The aquifers exploitation in coastal zones is generally a crucial issue since it combines the notion of quantity with quality (Ledoux, 1986). Coastal regions are often areas where there is generally a large demand for water due to the multiple activities of large agglomerations such as agriculture, industry and tourism. These overpopulated coastal areas with very intense economic activities, require quantity and uncontaminated water resources.

This coastal aquifer in contact with the sea is naturally in equilibrium, the groundwater coming from the infiltration of pluviometric water constitutes a groundwater moving towards the sea which surmounts a saltwater body in a wedge penetrating inside lands. Any over-pumping of freshwater from a wellfield will alter this equilibrium state by reducing the flow of groundwater and lowering the groundwater. This results in the extension of an inland salt wedge that can reach in some places, the bottom of the wells and even lead to a change in the direction of the runoff (Bonnet *et al.*, 1974; Gourgand *et al.,* 1988).

For example, the coastal areas (northern part) of the study area, where there is over pumping for domestic and industrial consumption, are areas where groundwater is vulnerable to salt contamination and where aquifers are in direct contact with the Mediterranean. The purpose of these studies is to investigate the mechanisms of seawater intrusion, to characterize saline pollution and to highlight the governing factors and the tools of controlling the contamination of water tables by seawater.

Freeze and Cherry (1979) define seawater intrusion as the migration of saltwater into aquifer freshwater, under the influence of the evolution of groundwater resources. The movement occurs naturally, either inland; this is a downward movement from the surface sources and irrigation water to the aquifer and an upward movement of the lower formations into the aquifer, or in coastal areas where aquifer waters are hydraulically linked to seawater (El Achheb *et al.*, 2001; Pulido-Bosch *et al.,* 1998).

In this latter case, the two systems do not mix, because saltwater move below freshwater owing to the density difference between the two types of water (Custodio and Llamas, 1983; Pulido-Bosch *et al.*, 1996; Martos and Pulido-Bosch, 1996; Hai Van and Sang-Il, 2015; Dilip Kumar and Bithin, 2017.

Many authors (Bear *et al.*, 2001, Calvache and Pulido-Bosh, 1991, Rivera *et al.*, 1990) have attempted, through analytical and numerical modeling methods, to describe the phenomenon, to predict the position of the interface between freshwater and seawater and predict the change of piezometric head and salinity.

Characterization of the intrusion phenomenon requires an interdisciplinary approach which consists in establishing the geological structure, determining the hydrological and hydrochemical properties by a sufficient number of measurements and analyzes in order to characterize the origins of the salinity, the location of the interface and the functions that cause spatio-temporal evolution (Banton and Bangoy, 1999). The captive aquifer studied in this article is located in the Seybouse basin in northeastern Algeria (Fig. 1 and 2).

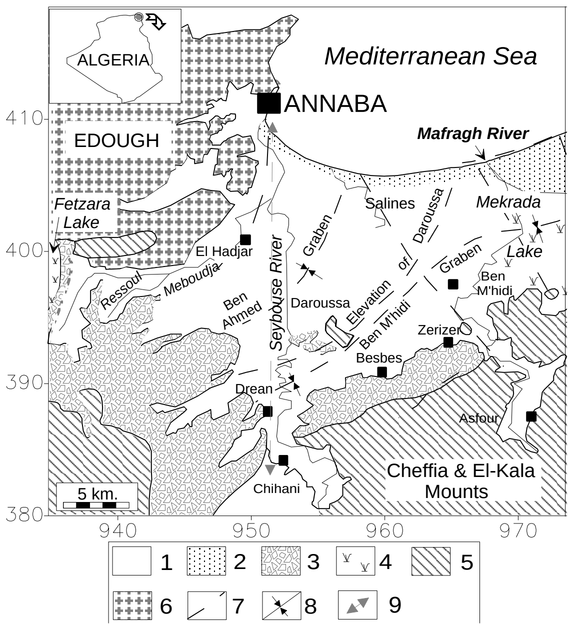

Fig. 1. Geological map of the Annaba plain

Legend: (1) recent alluvia; (2) dunes; (3) ancient alluvia; (4) lakes or ponds; (5) Numidian sandstones or clays; (6) metamorphic formations (micaschists, gneisses, marbles); (7) fault; (8) graben; (9) cross plot.

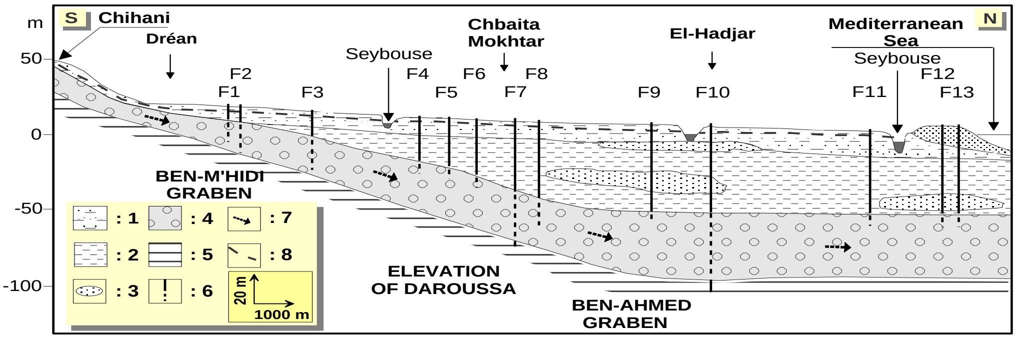

Fig. 2. Cross-section through the Annaba plain

Legend: 1: clayey sands (superficial aquifer); 2: detrital clays of Plio-Quaternary; 3: sands; 4: pebbles and gravels (deep aquifer); 5: Numidian clays; 6: boreholes; 7: flow path; 8: piezometric head of the deep aquifer.

The problem of this aquifer is due to the imbalance between recharge and overexploitation, the importance of agricultural activities and the presence of minerals with high solubility in its reservoir.

Material and methods

Mechanisms of seawater intrusion (Generalities)

The transition between freshwater and saltwater occurs relatively suddenly over certain thickness not exceeding a couple of meters. The two miscible liquids are so separated by a zone that is often assimilated to an abrupt interface limiting saltwater wedge, the slope of which is inclined towards the continent (Fig. 3).

The existence and the spatio-temporal evolution of the transition zone depend both on the hydrodynamic and geometric factors (Bonnet *et al.*, 1974; Ledoux, 1986):

✓ The natural variations both of piezometric head (seasonal variations) and the sea level (tide) that causes a mixing of freshwater and seawater by the interface movement;

✓ The density difference between the two liquids that tends to maintain salt in depth;

✓ The molecular diffusion of salt in freshwater that tends to reduce the concentration contrasts ($2.\ 10^{-9}$ m^2/s for the chlorides). It corresponds to the physicochemical dispersion (Schoeller, 1962; Castany and Margat, 1977);

✓ The (mechanical and cinematic) dispersion due to the flow along the interface; it is reflected by the formation of a mixing zone of freshwater and marine saltwater. It is quantitatively expressed by the intrinsic dispersion coefficients (longitudinal and transverse);

✓ The porosity and permeability of coastal aquifer;

✓ The aquifer geometry.

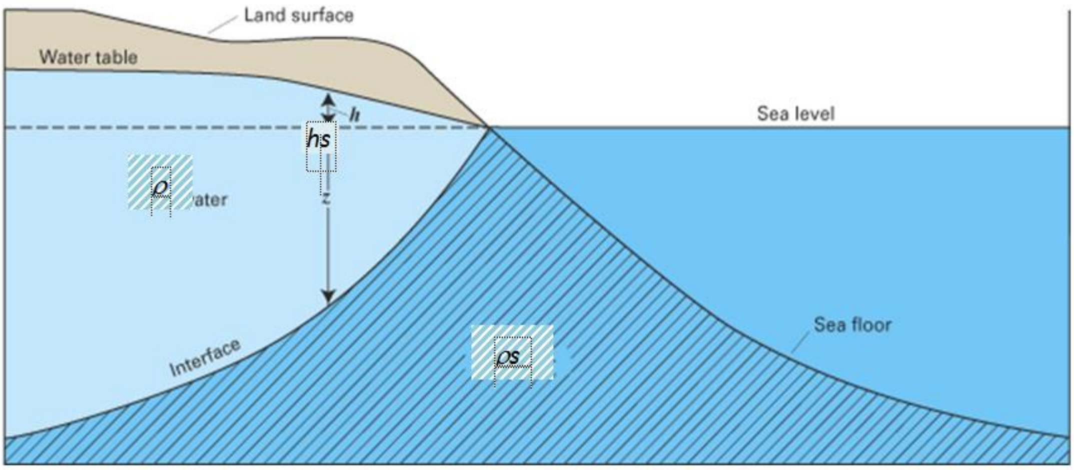

Fig. 3. Hydrodynamic graph of unconfined coastal aquifer according to Ghyben-Herzberg relation.

The water table discharge itself is function of the previous factors. The progressive invasion of the aquifer by the inflow of seawater depends on le the flow discharge of aquifer that tends to cause a permanent flushing of coastal aquifer.

Role of hydrodynamic factors and of exploitation

The explanation of the seawater intrusion phenomenon of coastal aquifers was made by Ghyben-Herzberg (1901); it is expressed by the relation between the freshwater load (h) above mean sea level and the depth (hs) of the freshwater-saltwater interface below sea level (Fig. 3).

The Ghyben-Herzberg equation is expressed as:

$$hs = h\frac{\rho}{\rho s - \rho}$$

Where ρ is the density of freshwater (1g/cm^3);

ρs is the density of saltwater (on average 1,025g/cm^3); hs is the depth of the wedge below sea level and h is the piezometric height measured from the sea level.

For these density values, the interface depth below sea level is expressed as:

$$hs \approx 40.h$$

This expression indicates that, according to the difference density of the two liquids, the interface position and depth are determined by the freshwater height above mean sea level (piezometric head of the water table).

However, this formulation should be used carefully because it supposes hydrostatic conditions and a permanent regime, rarely grouped in the nature. There are other formulations to study the seawater intrusion phenomena, we can for example state that of Todd (1980) that derives from the Darcy's law and is written as:

$$Q = 0{,}5\left(\frac{\rho s - \rho}{\rho}\right)K\frac{b}{L}$$

Where Q is the freshwater discharge flowing seaward (m^3/s);

K is the permeability of coastal aquifer (m/s);

b is the saturated thickness of unconfined aquifer;

L is the length of seawater intrusion in coastal aquifer.

In other words, this equation shows that the length of the saltwater wedge movement inland strongly depends on the aquifers terrains immediately in contact with the sea and on the power of the zone saturated with water. In contrast, it is inversely proportional to the discharge of groundwater runoff seaward.

Thus, the length of seawater intrusion into coastal aquifer is significant in the case of low discharge of groundwater and high permeability of the coastal sector. Conversely, in the case of a slightly permeable aquifer with a high discharge of groundwater runoff; i.e. with significant hydraulic gradients or large saturated thickness, the movement of saltwater toward the continent is low or even negligible. The previous equations indicated that the length of seawater intrusion depends on the discharge of groundwater runoff. In fact, any overexploitation in coastal areas above the underground reserves reduces the discharge of groundwater to the sea that constitutes its outlet and causes for the transitional zone freshwater-seawater to move inland.

On the whole, the discharge of the aquifer exploitation should equal the replenishment rate of the water table. An upwelling of a transition zone can appear even if the water table is not regionally exploited. This is a local upwelling of the interface between the two liquids beneath the wells in such a way that saltwater reaches the screens pipes: phenomenon called « up coning » (Bear *et al.*, 1999). This leads to a severe pollution of extracted water by the salts of seawater.

Methodology

Highlighting seawater intrusion requires an interdisciplinary approach. Many authors (Demirel, 2004; El Achheb *et al.*, 2003; Gemail *et al.*, 2004; Grassi and Cortecci, 2004; Kafri and Arad, 1979; Lebbe *et al.*, 1989; Paine, 2003; Pulido-Leboeuf, 2004; Spechler, 1994; Trabelsi *et al.*, 2005; Wilson *et al.*, 2006; Hani *et al.*, 2006), studied the phenomenon in order to locate the interface position between freshwater and seawater, using analytical, geophysical and modeling methods. Other studies (Allen and Suchy, 2001; Farber *et al.* 2004; Grassi and Cortecci, 2004; Martos *et al.*, 2001; Olobaniyi and Owoyemi; Pulido-Bosh *et al.,* 2003; Vengosh and Rosenthal, 1994) defined the processes and the chemical reactions that characterises the mineralization and that would be responsible for the enrichment or the impoverishment of groundwater in chemical elements.

The adopted procedure consists in:
- Studying the hydrodynamic factors that can play a key role;
- Determining the geochemical properties by measures and analyses to characterize the origins of salinization and the factors inducing its spatio-temporal evolution;
- Constructing concentrations crossed diagrams of major elements with Cl^- ion that constitutes a good salinity tracer;
- Finally, constructing a numerical model of pollutants transport.

Results and discussion

Highlighting seawater intrusion

Hydrodynamic factors

The coastal sectors where aquifers are in contact with the sea (north of the Annaba plain), are the most vulnerable to seawater intrusion that is enhanced by the following criteria (Chen et al., 1997):

✓ *Permeability:* in the gravel water table the highest permeability values lie along the Seybouse;

✓ *Thickness of gravels and pebbles:* it passes in fact from a couple of meters on the western edge of the system to nearly 25m to the north according to the axis of the Ben-Ahmed pit, oriented south-north, then it decreases to lower than 10 m on the Daroussa elevation. Between the Boukhadra and the El-Khous mound, another level with coarse elements was located at a depth of 35 to 40m (Djabri *et al.*, 2000);

✓ The geometric characteristics and the dip of gravels indicate that the aquifer ends at the sea many kilometers from the coast (Fig. 4);

✓ Low hydraulic gradients;

✓ Overpumping at the wellfields of the Salines and the Allelick causing a sharp lowering of piezometric heads up to -8m;

✓ Decrease in piezometric heads: the permanent extension of isopieze curve of 0 elevation caused by pumping reflects a generalization of seawater intrusion into freshwater via a transition zone. We particularly state the Salines and the Allelick depressions, the elevation of which can reach -10m and where the groundwater flow is directed from the sea toward the set of boreholes;

The monitoring of piezometric chronicles allows to highlight a generalized decrease of heads and to draw the following conclusions:
- The presence of some depressions with elevations below sea level and the general decrease in piezometric heads mainly lead to the elevation of freshwater-saltwater interface that can rapidly reach the bottom of the deep boreholes. We state the Salines and the Allélick depressions where the heads can reach more than 10 m;
- The permanent extension of the curve of zero elevation caused by the pumping reflects the generalization of seawater intrusion into freshwater via a transition zone;
- The sharp decrease in piezometric heads, especially in low water period and low seasonal variations of the water table leads to the reduction of freshwater discharge and to the penetration of seawater beneath the freshwater masses of the water table.

The hydrochemical study of seawater intrusion seems to be simple. However, this intrusion phenomenon of seawater is accompanied by other processes that modify the characteristics of water mixture.

This change is due to the absence of balance between the aquifer and the water mixture. In fact, carbonates and clays contribute to the dissolution and precipitation of some minerals and to cation exchange that behaves against changes caused by seawater intrusion.

Together with the reduction of sulfates, these processes are the modifying factors of hydrochemistry of water salinized by seawater intrusion (Petelas and Diamantis, 1999).

Fig. 4. Gravels geometry in the Annaba aquifer system.

Study of chemical analysis

Piper Diagram

By observing the diagram (Fig. 5), we notice that, for anions, the samples are rich in chlorides in their entirety. We notice that five samples constitute the mixed domain; i.e. that any anion dominates. By observing the distribution of cations, we notice a dominance of sodium; this latter is accompanied by potassium. It should be noted that a significant number of samples indicate an enrichment of water in calcium and magnesium. The combination of the two triangles indicates that water of the study area is classified as chloride-sodium type water and secondarily as calcium to magnesium type water.

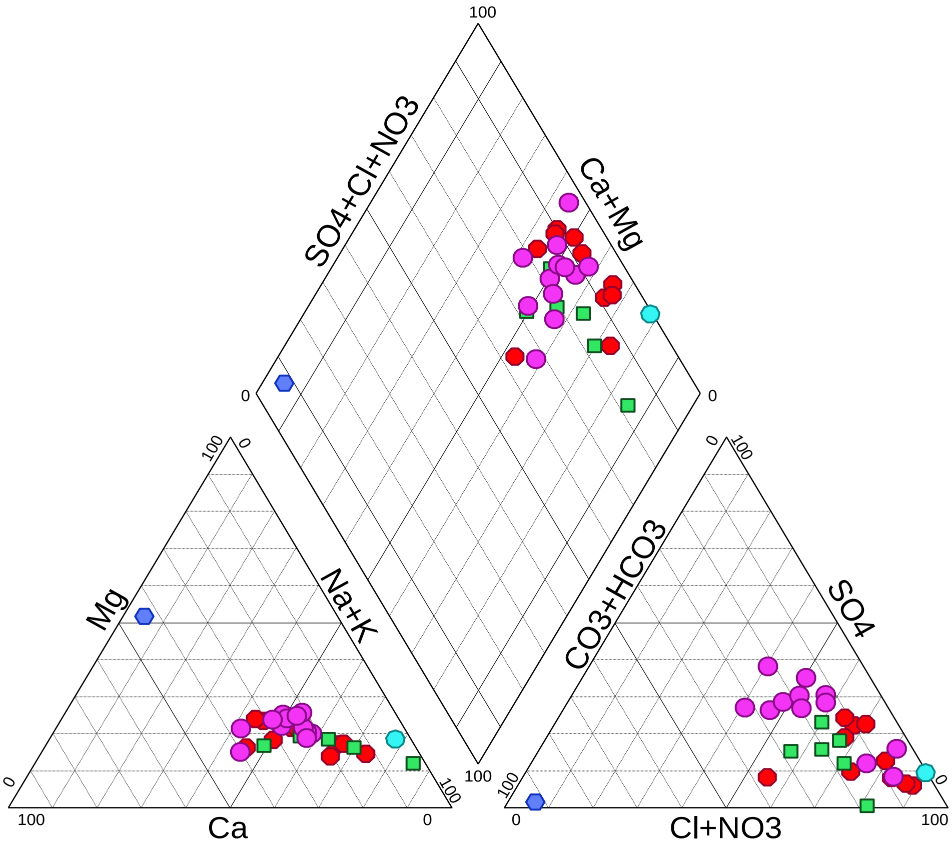

Fig. 5. Analyses results representation of water samples in Piper Diagram.

Stiff Diagram

The Stiff representation consists in constructing, for each sample, a diagram in the form of polygon that takes a geometric form according to the contents of the considered chemical elements (Fig. 6). The distinction between the samples is based on the polygon geometry that gives insight into the dominating species and the chemical relationship.

The three axes of Stiff Diagram are respectively, from top to bottom, Na-Cl, Ca-HCO_3, Mg-SO_4 (Fig. 7). The obtained Stiff Diagrams allowed dividing groundwater into three chemical homogenous classes: classes 1, 2 and 3.

- The class 1, accounting for nearly 70% of the total items, groups water of the coastal sector, at the Salines, Sidi Salem, El Hadjar and Ben M'hidi areas. The Stiff diagram of this class resembles that of seawater (Fig. 8);

- The class 2, grouping nearly 19% of the total items, characterizes the samples of the southern sector of the plain;

- The class 3, with 11% of the total items, is formed by the lowest mineralized water;

The spatial distribution of the different classes of Stiff Diagram allows understanding the Stalinization origin of groundwater.

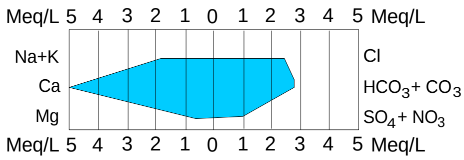

Fig. 6. Stiff Diagram of freshwater.

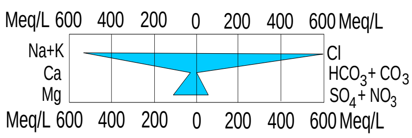

Fig. 7. Stiff Diagram of seawater.

Crossed diagrams of concentrations

The second tool of interpretation used in this study consists of crossed diagrams of concentrations together with chlorine ion. This latter as a conserved element, does not contribute to water-rock interaction and characterizes the origin of salinity of water constituting a mixing tracer (Tellam, 1995).

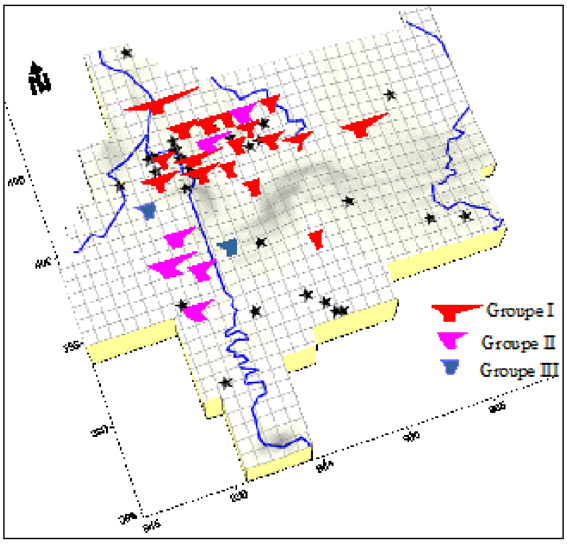

Fig. 8. Stiff Diagram of water of the Annaba aquifer (gravels aquifer).

The crossed diagrams show the relation between chlorides and major elements (Na^+, Mg^{2+}, Ca^{2+}, K^+ and SO_4^{2-}) of water points sampled at the study area. The disposition of the different points with respect to the mixing line freshwater-saltwater can be of great interest to identify other phenomena associated with the mixing process.

The diagram Na–Cl (Fig. 9) indicates that the representative points of the classes 1 and 2 are divided below and on mixing line of seawater-rainwater The points lying on the mixing line show the presence of the mixing process freshwater-saltwater without of ions exchange reactions (Kouzana *et al.*, 2007). The points of the classes 1 and 2 lie below the mixing line freshwater-saltwater indicating an impoverishment in sodium (El Achheb *et al*, 2003). These waters are especially controlled by cation exchange reactions. Given that Na^+ content should balance Cl^-content, the Na^+ deficit is explained by the reverse ion exchange phenomenon between water and aquifer and reflected by Na^+ adsorption and Ca^{2+} release (El Achheb *et al.*, 2003).

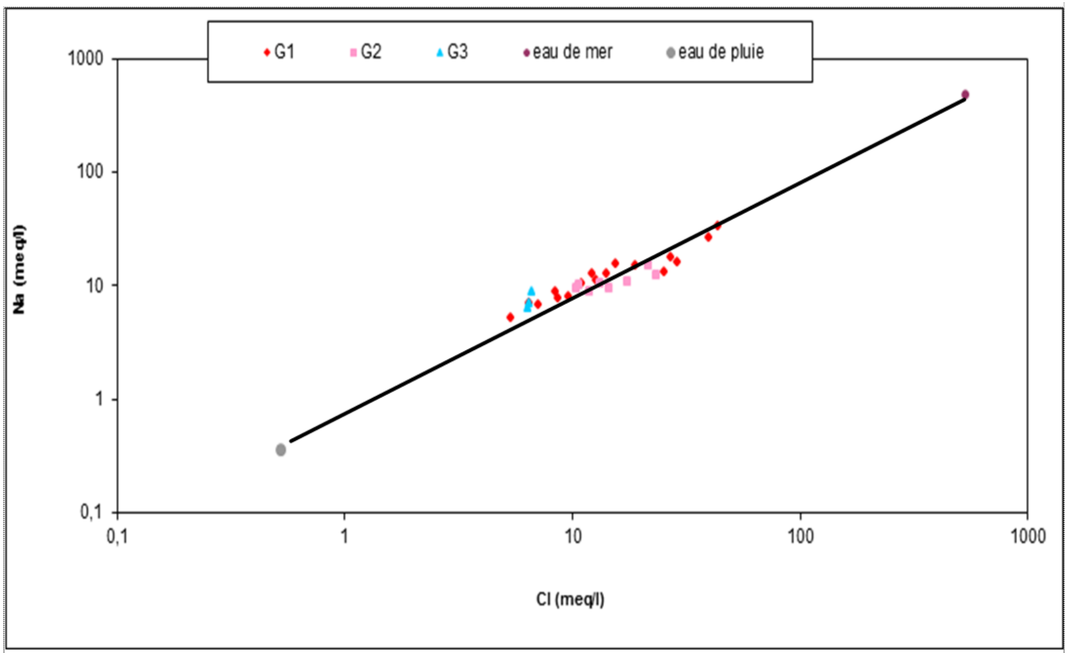

Fig. 9. Relation between groundwater Na-Cl and that of seawater.

The Ca–Cl diagram (Fig. 10) is a good illustration of data obtained from Na-Cl diagram, showing a general enrichment in calcium with respect to the mixing line seawater-rainwater. The amplitude of this increase is different from one class to another.

The class 2 and 3 exhibit the maximum enrichment and the class 1 the minimum. The points of graph Mg-Cl (Fig. 11) lying below the mixing line are characterized by the impoverishment in Mg^{2+}. This is especially explained by the phenomenon of water-rock interaction.

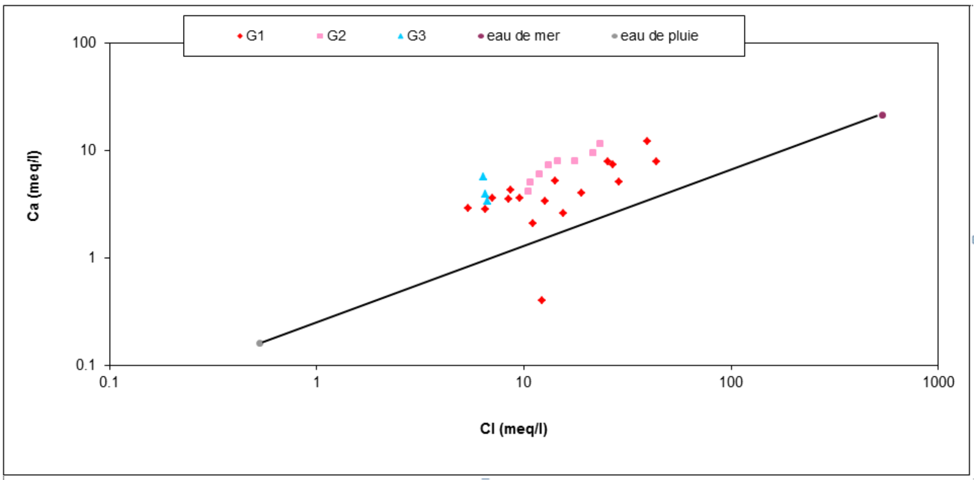

Fig. 10. Relation between groundwater Ca-Cl and that of seawater.

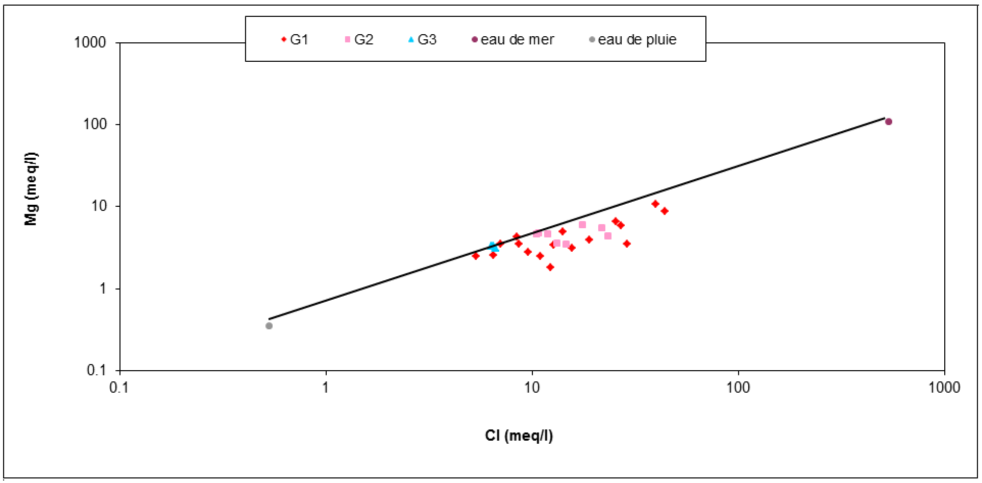

Fig. 11. Relation between groundwater Mg-Cl and that of seawater.

The diagram NO3–Cl (Fig. 12) shows that the increase in nitrates is accompanied by an increase in chlorides, particularly for the samples of class 1 and 2, indicating enrichment owing to the movement of irrigation return flow.

This latter movement coupled with the evapotranspiration facilitates the minerals dissolution process and causes the dissolution of the agricultural fertilizers and residues, thereby leading to a decrease in Ca, HCO_3 and SO_4, as a result of precipitation, and an increase in Na, Cl and NO_3 (Cardona *et al.*, 2004; Richter and Kreitler, 1993).

The relation between $SO4^{2-}$ and Cl^- (Fig. 13) shows that the totality of the points are below the straight line of mixing freshwater-saline water except for two point of the group III, the sulphate comes from the dissolution of the irrigation waters the area. Reduction processes would explain the SO4 deficit for Group I and II waters (El Achheb *et al.,* 2001).

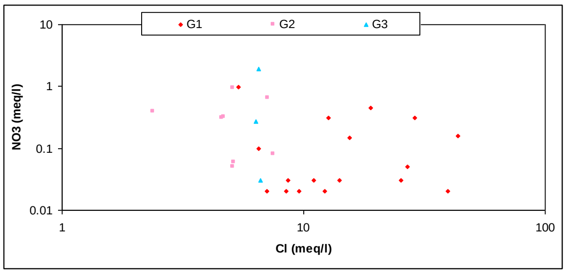

Fig. 12. Relation between groundwater NO_3-Cl and that of seawater.

The Mg^{2+}/Ca^{2+}ratio increases according to the seawater proportion intruded in the mixture (Tellam, 1995; Trabelsi *et al.*, 2005). The enrichment in magnesium and the impoverishment in calcium are due to ion exchange reactions typical of the mixing movement freshwater-seawater.

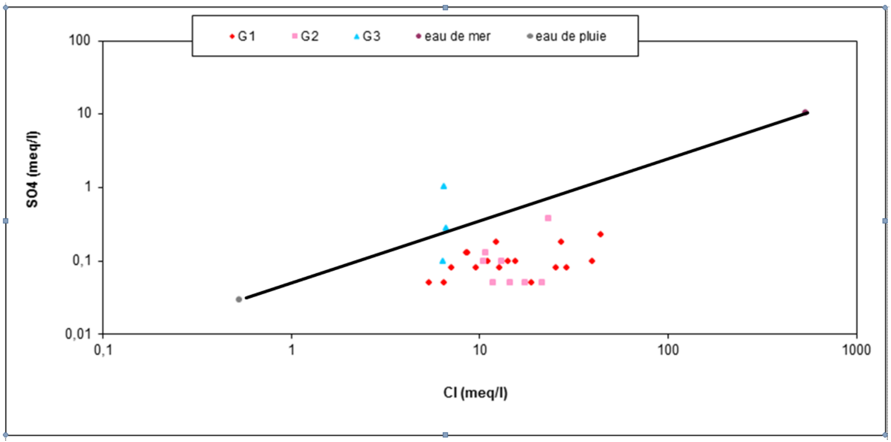

Fig. 13. Relation between groundwater SO_4-Cl and that of seawater.

In the Fig. 14, the points of class 1 exhibit the highest values of this ratio, proving the seawater origin of mineralization of these waters. one part of the points of class 2 lies on the mixing line rainwater-seawater, thereby confirming the phenomenon of irrigation returns; the other part of the points of class 1 as well as the points of class 3 lie below the mixing line exhibiting the lowest values of Mg/Ca ratio because of the enrichment in calcium ion deriving from gypsum and calcite present in the reservoir. The SO_4^{2-}/Cl^-ratio decreases when the seawater proportion (represented by the amount of chlorides) increases in the freshwater-seawater mixture (Pulido-Bosh *et al.*, 2003; Trabelsi *et al.,* 2005; Vengosh and Rosenthal, 1994). The Fig. 15 shows that the points of class 1 exhibit the lowest values of the ratio, confirming the marine origin of water. In contrast, the points of class 3 exhibit very significant values of SO_4^{2-}/Cl ratio, indicating another origin of mineralization; we think that this ratio increase of class 1 points is due to gypsum dissolution in the reservoir (Trabelsi *et al.,* 2005).

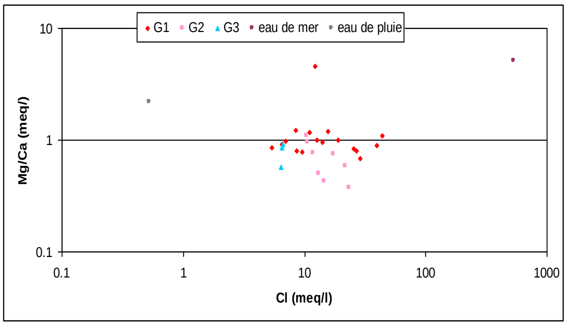

Fig. 14. Variation in $Mg2^+/Ca^{2+}$ ratios according to contents of chlorides (meq/l).

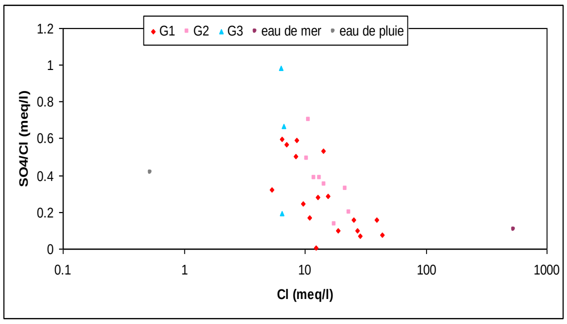

Fig. 15. Variation in SO_4^{2-}/Cl^- ratios according to contents of chlorides (meq/l).

Mineralization evolution of groundwater according to distance from the sea

In order to highlight the influence of distance from the coast on groundwater amount, here we review the analyses of groundwater samples performed according to three profiles S-N perpendicular to the limit with the Mediterranean (Debièche, 2002; Aoun-Sebaiti, 2003).

The graphs of Fig.s 16 a-f show a sharp drop in values for the whole chlorides, sodium and electrical conductivity. This drop is very sharp in the first 15 kilometers of the coast. In this sector, the values of chlorides passe from 800mg/l to lower than 200 mg/l and the values of sodium drop by 400mg/l to lower than 100mg/l. Finally, conductivity decreases by near 3500 to 1500µS/cm. The values significantly go back up again southward for the different dosed elements. The increase of strontium values (Fig. 16 g-h), in this sector, may reflect the influence of evaporitic formations on the physicochemical content of water (Debièche, 2002; Djabri *et al.*, 2003; Aoun-Sebaiti *et al.,* 2013).

The profile 2 (Fig. 16), grouping water points perpendicular to the sea and taking up the center of the plain, shows a joint decrease in Cl, Na and electrical conductivity, moving away from the sea over a distance of near 2,85km. But, unlike the first profile going along the Seybouse, this one shows, after a stabilization of contents, a decrease in values, moving southward. This is explained by the absence of leaching of evaporitic formations conveyed by the Seybouse. The profile 3 located east of the two first ones shows similarity to the second one, however, with much lower contents (Fig. 16). High salinity, around 450 mg/l of chlorides, is observed in a sector extending up to 5, 4km inland. The contents drop suddenly southward, probably for the same reasons than it is for the second profile. It is obvious that highlighting seawater intrusion implies the multiplication of monitoring of many parameters (CE, Cl^-, Na^+, Br^-, O^{18}, and…) on several profiles perpendicular to the sea and parallel to the current lines (Mania *et al.*, 1985 ; Younsi *et al.*, 1997; Younsi, 2001). In the Annaba plain, we realized profiles, where we have a number of sufficient points of observation. In the other sectors, the measures are fewer and do not allow to draw a significant profile.

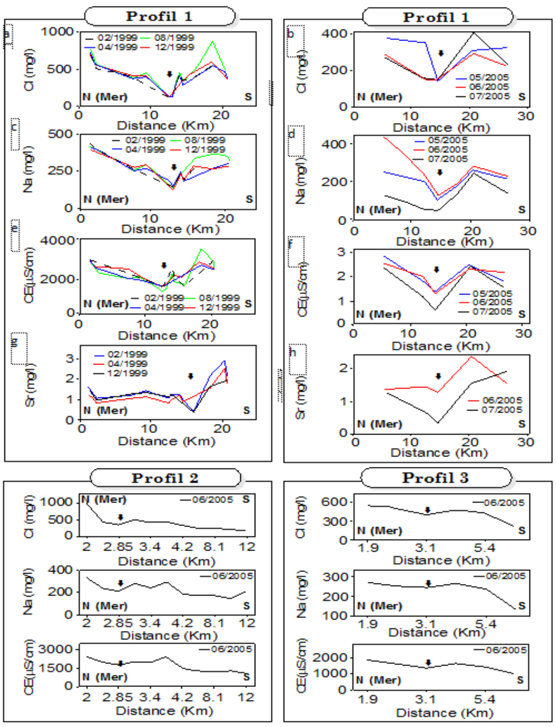

Fig. 16. Temporal evolution of mineralization according to 3 profiles orthogonal to the sea.

Conclusion

The contamination problem of the gravels water table by seawater is caused and worsened by overpumping of wells of the northern part of the plain to meet the agricultural, industrial and domestic needs of a growing population. This overexploitation caused an increase in piezometric head, thus allowing for a saltwater wedge to progressively intrude into groundwater.

To highlight this phenomenon, we resorted to several hydrochemical methods that revealed this:

- From Piper Diagram we could observe the dominance of chlorides and sodium in the entirety of samples, thus revealing a chloride-sodium facies.
- Stiff diagram, for its part, divides groundwater into three classes: the first class (class 1), the form of which resembles seawater, represents coastal groundwater at the Saline and Sidi Salem and at El Hadjar and Ben M'hidi, the second class (class 2) characterizes the samples of the southern sector of the plain, the third class (class 3) represents slightly mineralized water.
- The crossed diagrams of concentrations that show the relation between Cl and the other major elements with respect to the mixing line rainwater and seawater also explain:

• The phenomenon of ion exchange between the rock and water and which is reflected by the groundwater impoverishment in Na and an enrichment in Ca and Mg.

• The increase in NO_3 in water of class 1 and 2 reflects the irrigation return flow in this zone.

The different ratios studied in these diagrams also show:

• The water mineralization of class 1 exhibiting the highest values of Mg/Ca ratio seems to be marine, whereas it seems to derive from gypsum and calcite present in the reservoir for the class 3 and a part of class 3 points.

• SO_4^{2-}/Cl^- ratio seems to confirm the results of Mg/Ca ratio for the water mineralization origin of the three classes of water.

- The temporal mineralization of the gravels phreatic surface water according to three profiles parallel and perpendicular to the sea also indicates a sharp drop in electrical conductivity and Na, Cl concentrations, as one goes away from the sea, thus seemingly confirming the seawater assumption.

Acknowledgments

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