



RESEARCH PAPER

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Hydrogeochemical characterization and assessment of the quality of surface waters of Oued Saf-Saf (North-East Algeria)

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Article published on February 28, 2018

Key words: Oued Saf-Saf, Hydrogeochemistry, Pollution parameters, Surface water

Abstract

The study of the hydrogeochemical characteristics for evaluation of surface water quality in the Oued Saf-Saf required the taking of 13 surface samples all along the watercourse, from upstream to downstream in order to get an idea of the evolution of water quality on the route from the wadi to the mouth of the Mediterranean Sea. The analyzes therefore focused on the determination of the principal mineral elements, chemical and biochemical oxygen demand, lead, copper, total coliforms, fecal coliforms, fecal streptococci and clostridium sulphito-reducers. Examination of the variation in the chemical composition of the surface water in the Oued Saf-Saf along the flow line to the outlet indicates high concentrations of bicarbonates and calcium probably at the dissolution of calcic and carbonate facies. The denitrification process, the dissolution of dolomite and calcite can explain the geochemical characteristics of the surface waters studied. The determination of organic pollutants by the organic pollution indices (IPO) and the biodegradability index (IB) is therefore based on the parameters of BOD₅, COD, O₂, nitrates, nitrites, ammoniums, phosphates and the microbiological quality index (IQM). The results obtained describe a worrying situation of the state of this river, dangerously threatened by human activities. The IPO shows low to moderate pollution that changes from upstream to downstream and the IB is greater than 3, indicating non-biodegradable effluents. On the other hand, the IQM suggests a moderate fecal contamination, which can be explained by the continuous improvement of the connection of the riverins to the collection and seeding networks.

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Introduction

Today, the major concerns of developing countries is to try to define the most accessible means to limit the different pollution generated by new activities imposed by the development of the sectors of agriculture, industry and urbanization accelerated by strong population growth (Chaffai *et al.*, 2006). Uncontrolled industrial discharges, fertilizers and chemicals in the agricultural sector and demographic pressure on water demand only increase the vulnerability of the receiving environments and the degradation of surface and groundwater resources in particular and the environment in general (Sakaa *et al.*, 2013). These different forms of pollution produce a chemical modification of the water and make it unsuitable for the desired uses (Khelfaoui *et al.*, 2009). Saf-Saf is one of the wadis of the surface network of the city of Skikda, currently threatened by human activities (domestic, urban, industrial waste and the use of water for agricultural needs, etc.). On the banks of the wadi Saf-Saf there are 10 villages, of which only four are equipped with sewage treatment plants with a treatment capacity of 42000m³/day. The volume of untreated wastewater is approximately 16153.2 m³/day.

The deterioration of water quality in the wadi Saf-saf region would lead to the loss of the natural heritage of the city and the contamination of groundwater and the environment if protection measures are not recommended (Sakaa *et al.*, 2015, Khelfaoui *et al.*, 2014).

In light of all these elements, the main objective of this study is to evaluate the hydrogeochemical processes of surface water and the state of pollution reached in this region of Saf-Saf.

Materials and methods

Study zone

According to the Hydrographic Basin Agency (ABH, 2000), the watershed of the wadi Saf-Saf is part of the Constantine coastal basin, between the watershed of Wadi Guebli in the West and that of the wadi Kebir West at the eastern part. It is bordered to the North by the Mediterranean Sea, to the East by Jebel Alia and Jebel Tengout, to the South by Jebel Hadja and Jebel Oucheni and to the West by the Collo Massif and Jebel Boukhalouf. It is registered in the territory of Skikda and extends to the territory of Constantine. The wadi Saf-Saf drains an area of about 1158 km² and empties east of the city of Skikda in the Mediterranean Sea (Fig. 1).

Geological overview

The region of the wadi Saf-saf is located in the small Kabylie which is part of the alpine chain of the Maghrebides in Algeria. It stretches between the Babors Massif in the West and the Edough Massif in the East for more than 150 kilometers along the coast. The Maghrebian chain shows from north to south five domains: the inner domain, the flysch domain, the outer domain, the eruptive rocks and the Quaternary

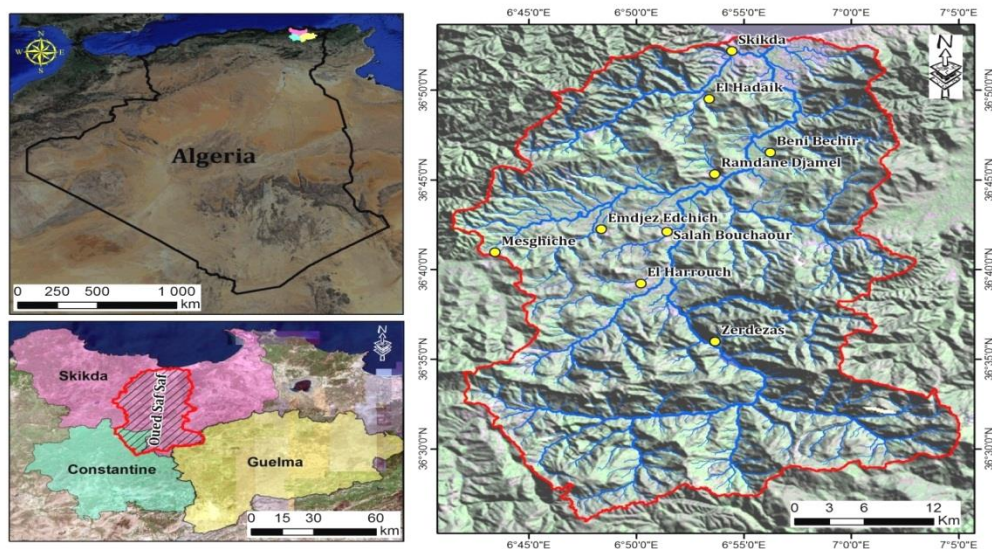


Fig. 1. Location map of the watershed of the wadi Saf-Saf.

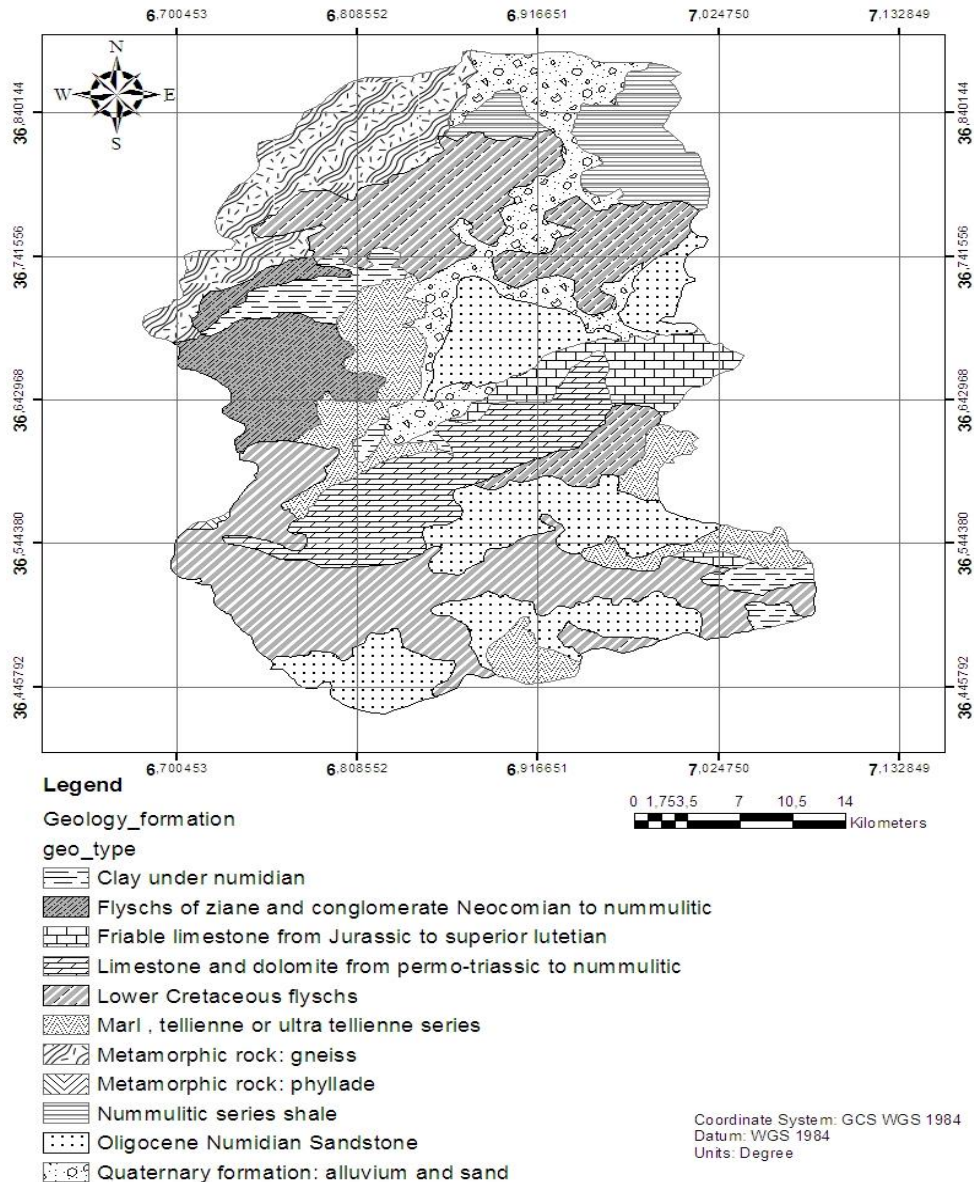


Fig. 2. Lithological map of the Saf Saf Valley.

The inner domain is composed of crystallophyllian metamorphic massifs or a Paleozoic sedimentary ensemble of Ordovician to Carboniferous age that is poorly metamorphic or unconformably covered by detrital deposits consisting mainly of late Oligocene, Early Miocene conglomeratic molasses (Mahjoub and Merle, 1990).

The domain of flyschs from Cretaceous to Eocene age, is subdivided into three types of flyschs (Mauritanian, Massylian and Numidian). The outer domain, also called the Tellian domain, of Middle Cretaceous to Neogene age consisting mainly of marl (Colom *et al.*, 1954).

The calcareous-type Miocene eruptive rocks, which generated large masses of granite (Bougaroun granite, Beni Touffout and Fila fila) and microgranite around Collo and west of the basin. Tamalous as well as acid lava flows and injections in the region of Collo, Chetaibi and Edough (Raoult and Mousterde, 1969). Ancient alluvial deposits and dunes are the two most dominant quaternary formations in the study area.

Situation of samples and analysis data

Sampling was carried out on the banks of Oued Saf-Saf from the upstream to Skikda town downstream (Fig. 3). The results of the 13 samples analyzed are shown in Table 1.

Table 1. Analysis data taken along the Oued Saf-Saf.

Parameters	T°C	pH	Cond. 25°C µS/cm	HCO ₃ ⁻	Cl ⁻	SO ₄ ⁻	Ca ⁺⁺	Mg ⁺⁺	Na ⁺	K ⁺	NO ₃ ⁻	NO ₂ ⁻	NH ₄ ⁺	PO ₄ ⁻	P total	O ₂ diss.	Cu	Pb	BOD ₅	COD
Sampling area	(mg/l)																			
Zardazas Dam	23,7	8,1	670	170,8	17,75	33	60,2	7,08	8,97	0,006	3,2	0,05	0,1	0,05	0,15	1,09	0,31	0,081	2	32
Zerdazas 1	23,6	7,7	820	225,7	35,5	34	81,76	27,72	34,73	0,036	6,3	0,06	0,15	0,08	0,12	1	0,23	0,053	3,5	34,4
Zerdazas 2	24	7,8	820	231,8	35,5	32	80,16	33,48	6,21	0,006	4,9	0,07	0,2	0,08	0,18	1,19	0,68	0,065	1,5	35
Said Bousbaa 1	23,8	7,8	790	213,5	53,25	36	60,12	23,88	17,25	0,011	3,6	0,05	0,32	0,23	1,78	0,96	0,38	0,093	5	35,3
Said Bousbaa 2	23,6	8	880	237,5	63,9	32	72,14	28,68	8,51	0,008	5,2	0,05	0,35	0,25	0,98	1,3	0,72	0,096	3	36,1
El-Harrouch 1	23,4	8	1180	292,8	124,25	34	84,17	47,88	23,23	0,032	3,9	0,06	4	0,45	7,8	2,3	0,55	0,069	1,5	39
El-Harrouch 2	23,2	7,6	1040	268,4	88,75	37	80,16	38,28	15,41	0,009	5,9	0,08	4,12	0,56	6,9	2,19	0,14	0,078	2,5	38,6
Salah Bouchaour 1	23,7	7,5	1180	268,4	106,5	39	80,16	40,68	18,63	0,028	4,1	0,06	3,44	6,85	0,53	1,4	0,71	0,095	2	51,4
Salah Bouchaour 2	23,4	7,6	1180	305	106,5	37	84,17	35,88	36,11	0,036	3,5	0,04	3,47	6,12	0,7	1,46	0,19	0,083	3	50,9
Ramdan Djamel 1	23,4	7,5	1280	329,4	124,25	35	92,18	40,68	37,49	0,039	3,7	0,02	4,52	1,17	7,85	1,3	0,41	0,072	5	55
Ramdan Djamel 2	23,4	7,6	1270	305	134,9	36	92,18	38,28	40,25	0,056	3,5	0,05	4,13	2,17	6,95	1,9	0,21	0,071	5	54,6
Hamadi Krouma	23,6	8,1	1170	286,7	124,25	33	88,18	43,08	20,47	0,029	4,2	0,05	6,2	5,41	2,28	1,13	0,85	0,114	1,5	61,8
Skikda	23,7	7,7	1020	256,2	88,75	42	76,15	40,68	8,74	0,007	6,25	0,04	5,9	4,87	2,67	1,23	0,56	0,106	0,5	86

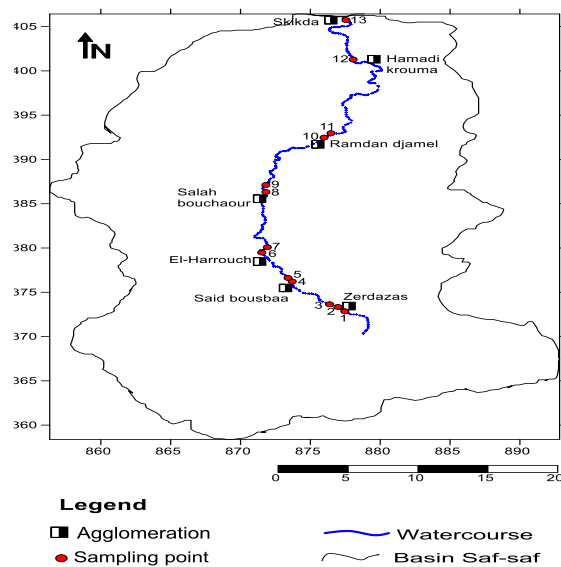


Fig. 3. Location map of analyzed samples.

Results and discussions

Geochemical facies

Given the predominance of these ions (HCO₃⁻, Ca⁺⁺, Cl⁻ and Mg⁺²) in surface water, a representation of the geochemical facies in a Ca⁺⁺/Mg⁺² and Cl⁻/HCO₃⁻ diagram proves useful. The diagram shows the existence of a single characteristic pole of water (bicarbonate-calcium facies)(Fig. 4).

Saturation index

The use of the Debye-Huckel law (1923) allows us to calculate the saturation index: IS= [log (Q)]/[log (Kps)] of minerals dissolved in water. The equilibrium state of this index is in the range of -0.5 to +0.5 (Table 2).

Table 2. Variation in the saturation indices of some minerals and the partial pressure of CO₂.

Sample	Aragonite	Calcite	Dolomite	Gypsum	Anhydrite	Halite	Sylvite	Cérusite	Anglesite	Log (PCO ₂)
1	0.57	0.71	0.83	-2.09	-2.4	-8.35	-11.07	-0.54	-4.79	-2.89E+00
2	0.38	0.53	0.92	-2.04	-2.36	-7.48	-10.01	-0.72	-4.74	-2.38E+00
3	0.49	0.64	1.23	-2.08	-2.39	-8.23	-10.8	-0.63	-4.8	-2.47E+00
4	0.34	0.48	0.89	-2.11	-2.43	-7.6	-10.35	-0.47	-4.52	-2.50E+00
5	0.64	0.79	1.51	-2.12	-2.43	-7.84	-10.41	-0.46	-4.83	-2.65E+00
6	0.73	0.88	1.84	-2.1	-2.42	-7.12	-9.53	-0.61	-5.05	-2.58E+00
7	0.3	0.45	0.9	-2.05	-2.37	-7.44	-10.22	-0.54	-4.51	-2.22E+00
8	0.23	0.38	0.8	-2.03	-2.35	-7.28	-9.66	-0.47	-0.63	-2.12E+00
9	0.4	0.54	1.05	-2.04	-2.36	-7	-9.54	-0.52	-4.56	-2.17E+00
10	0.34	0.49	0.94	-2.05	-2.37	-6.92	-9.45	-0.58	-4.58	-2.04E+00
11	0.41	0.55	1.05	-2.03	-2.35	-6.85	-9.25	-0.59	-4.63	-2.17E+00
12	0.86	1.01	2.04	-2.09	-2.4	-7.18	-9.58	-0.4	-4.96	-2.68E+00
13	0.38	0.53	1.11	-2.02	-2.33	-7.69	-10.33	-0.42	-4.42	-2.33E+00

Concentration diagram

We plot the analyzes in concentration diagrams by the logarithm of the molalities (meq.l⁻¹) on the ordinate and the logarithm of the abscissa concentration factors. The water concentration factor (FC) can be estimated from the ratio of the Cl⁻ content of a water

to the minimum value of Cl⁻ content in the database. This will allow a relative ranking of the samples relative to each other (Barbière *et al.*, 1992). The chloride ion (Cl⁻) is a good indicator of the concentration state of water, since this ion precipitates only at higher ionic strengths.

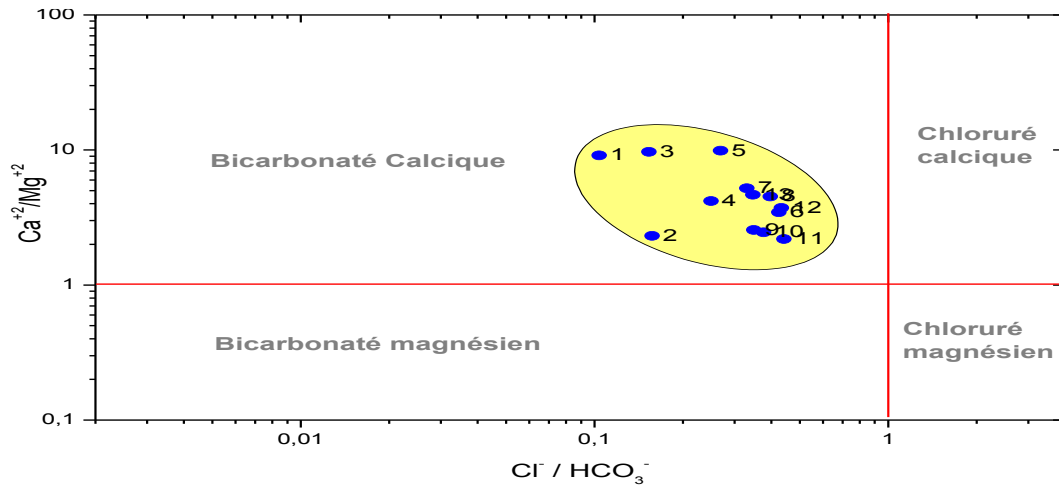


Fig. 4. Geochemical facies of surface waters (Reports: $Ca^{+2} / Mg^{+2} - Cl^{-} / HCO_3^{-}$).

Diagram of concentration of major elements

The carbonate facies is dominant in the surface waters sampled from Oued Saf-saf. The concentration of the carbonate alkalinity in the waters of the region can come from the dissolution of limestones or dolomites present both in the study area. It is noted that the carbonate alkalinity evolves with the global concentration and seems to be taken from the solution to participate in dissolutions from the beginning (Fig. 5).

As the waters concentrate, the calcium contents increase, the alkalinity (Alc) increases and then decreases according to the law of residual alkalinity (Tardy, 1980). As long as the waters are below saturated with respect to calcite, Ca^{+2} and Alc can increase simultaneously. When equilibrium with this mineral is reached, the Ca^{+2} content increases and the alkalinity decreases because $Ca^{+2} > Alc$, residual alkalinity positive calcite (Rodier, 1984).

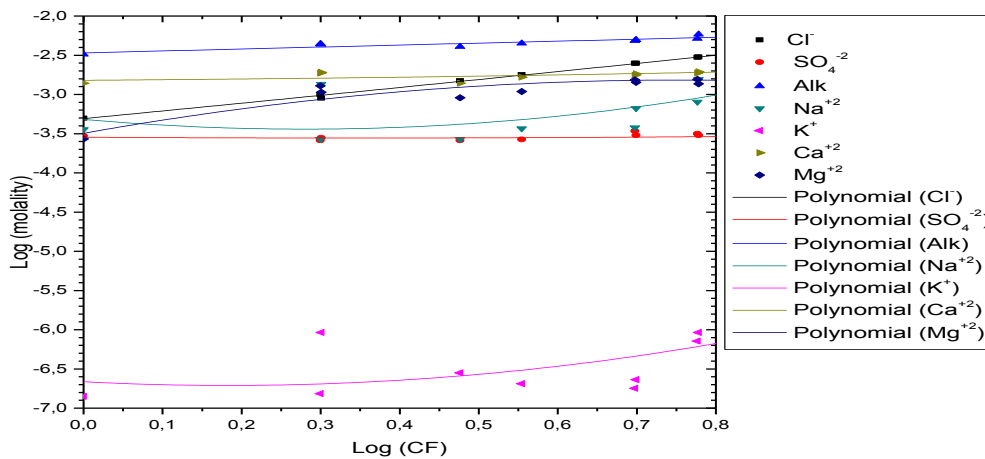


Fig 5. Concentration diagram of major elements.

Calcium is less important than carbonates but it is marked by a faster evolution than the concentration factor at the beginning, then the evolution of the concentration factor becomes more important than it. The importance of calcium ion compared to bicarbonates indicates an excess of the latter in the waters of the region, probably due to an origin other than carbonates (gypsum for example).

Sodium is marked by a significant evolution with the concentration factor. This can be explained by a simultaneous dissolution of chlorides and sodium from the same origin; we are talking about halite which is marked by a saturation index between -8.35 and -6.85, thus under-saturated. The Mg^{+2} content increases but slightly less than the concentration factor and the sulphates remain almost stable.

This result suggests a slight adsorption of Mg^{+2} by the humic clay complex and therefore an increase in the Ca^{+2}/Mg^{+2} adsorbed ratio (Ouardi *et al.*, 2014).

Potassium shows very low concentrations. However, it is noted by a significant increase in some samples (participation in probable precipitation of Sylvite (KCl) and a contribution of this element which increases its concentration) and a decrease for other samples. In general, the evolution of potassium remains very slow as the concentration factor.

Nutrient concentration diagram

Fig. 6 shows a perfect evolution of NH_4^+ ammonium salts, ammonia NH_3^+ and phosphate ion PO_4^{-3} with the concentration factor apart from a few samples. Phosphates are considered to be the main cause of eutrophication of surface water. This importance PO_4^{-3} concentration is mainly due to the supersaturation hydroxyapatite marked with a saturation index between 1.86 and 8.22. In view of the high concentration of NH_3^+ and the perfect evolution of CO_2 (Fig. 7), the studied surface waters become poisoned (Fig. 6).

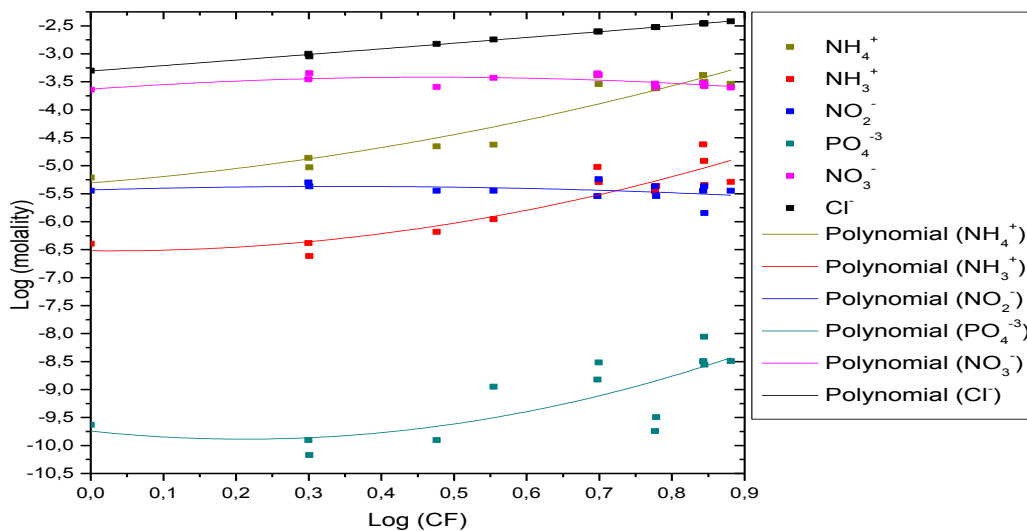


Fig 6. Nutrient Concentration Diagram.

For nitrates NO_3^- and nitrite NO_2^- , there is a decrease in the evolution of the latter with the concentration factor. For nitrites and ammonium, their presence in water is linked to the denitrification process by oxygen consumption. The ammonium concentration increases when the water contains a high CO_2 content and a low O_2 content as in the case of Fig. 7. In an oxidizing medium, the ammonium is converted into nitrites and nitrates; which induces oxygen consumption (Gaujous, 1995). Ammonium is the most toxic form of nitrogen and the presence of ammonium in large quantities is an indication of contamination by releases of human or industrial origin.

Concentration diagram of carbonates and metals

The content of bicarbonates HCO_3^- increases more rapidly than the concentration factor. This is due to the dissolution of limestones and dolomites and the

dominance of the carbonate facies of surface waters. The evolution of CO_3^{2-} ion and due mainly to the presence of the latter in aragonite $CaCO_3$ compounds, Calcite $CaCO_3$ and dolomite $CaMg (CO_3)_2$ which are marked by saturation supersaturated index (Table 2). Carbon dioxide CO_2 is marked by an evolution with the concentration factor. This evolution depends either on the decomposition of dead plants and fish that are reduced by bacteria, or on the respiration of fish that release a lot of CO_2 . With regard to metals, copper (Cu) shows a decrease in the evolution with the concentration factor, but unfortunately it is not the case for lead (Pb) which is marked by a tangible evolution due certainly at the dissolution of Cerusite ($PbCO_3$) and Anglesite ($PbSO_4$). Lead is a toxic substance and there is no threshold under which exposure to lead would be safe (Fig. 7).

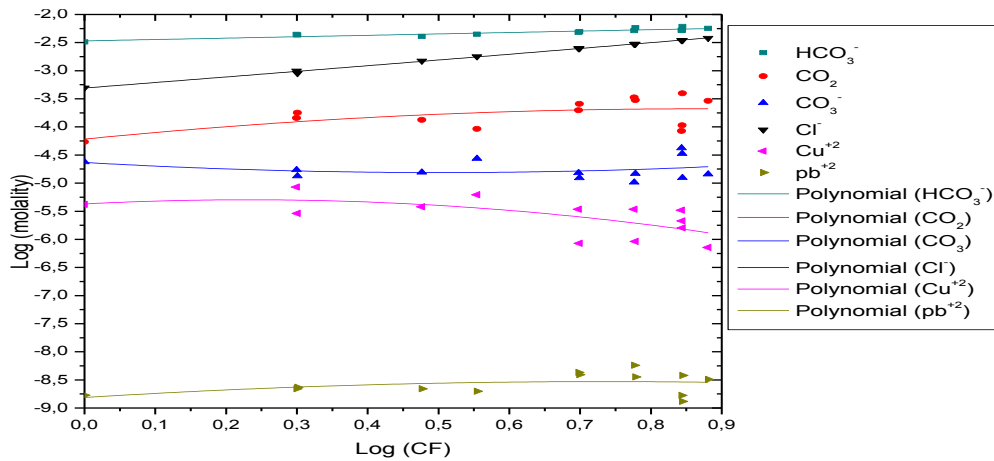


Fig 7. Concentration diagram of carbonates and metals.

Water-calcite balance diagram

The pCO₂ of river waters is generally in equilibrium with the atmosphere, whereas a higher pCO₂ (Log pCO₂ > -1.5) is the result of a deep circulation of groundwater, which had an interaction with carbonates (Thilagavathi *et al.*, 2012).

It is noted that calcite supersaturation increases when pCO₂ decreases (Fig. 8), this phenomenon has already been observed in the literature (Dosso, 1980 and Valles, 1987)(Fig. 8).

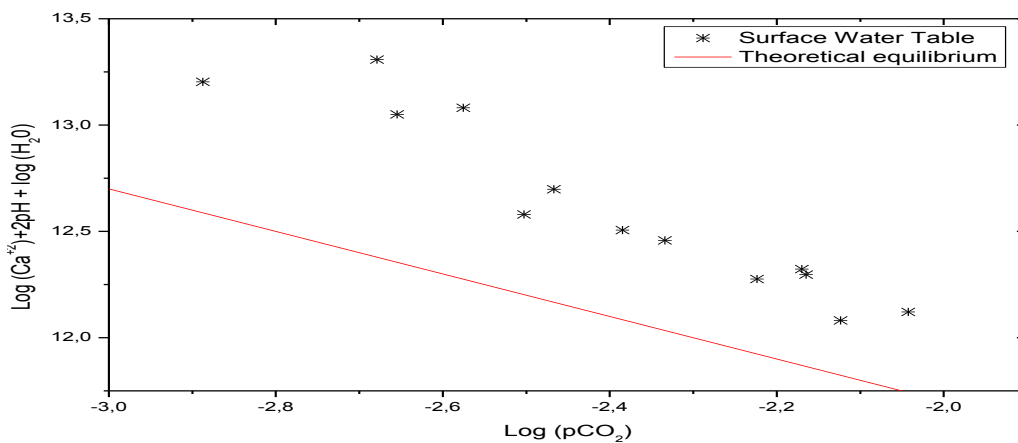


Fig. 8. Water-calcite balance chart.

Stability index of carbonate minerals by pH

The stability indices of the main carbonate minerals (Aragonite, Calcite and Dolomite) show that the latter are at equilibrium. This state of carbonate minerals indicates that the water has undergone significant changes as a result of CO₂ degassing (Fig. 9).

positive linear trend, which indicates that the mineral phases are changing with a similar trend following the same hydrogeochemical process (Fig. 9b).

This induces a precipitation of some of the carbonate minerals (Djidi *et al.*, 2008). The saturation indices of Calcite, Aragonite and Dolomite increase steadily with pH (Fig 9a). The alkalinity produced by the dissolution of carbonates is the main cause of the increase in pH. The SI of these minerals follows a

Relation Log (Pco2) vs IS calcite and IS dolomite:

In order to provide details on the hydrochemical processes of the study area, a use of the partial pressure of carbon dioxide (PCO₂) associated with that of the pH and the saturation index of Calcite and Dolomite is often very necessary. The log (PCO₂) and IS calcite and log (PCO₂) and IS dolomite bonds show that all the samples are supersaturated and that they are both precipitating (Fig. 10).

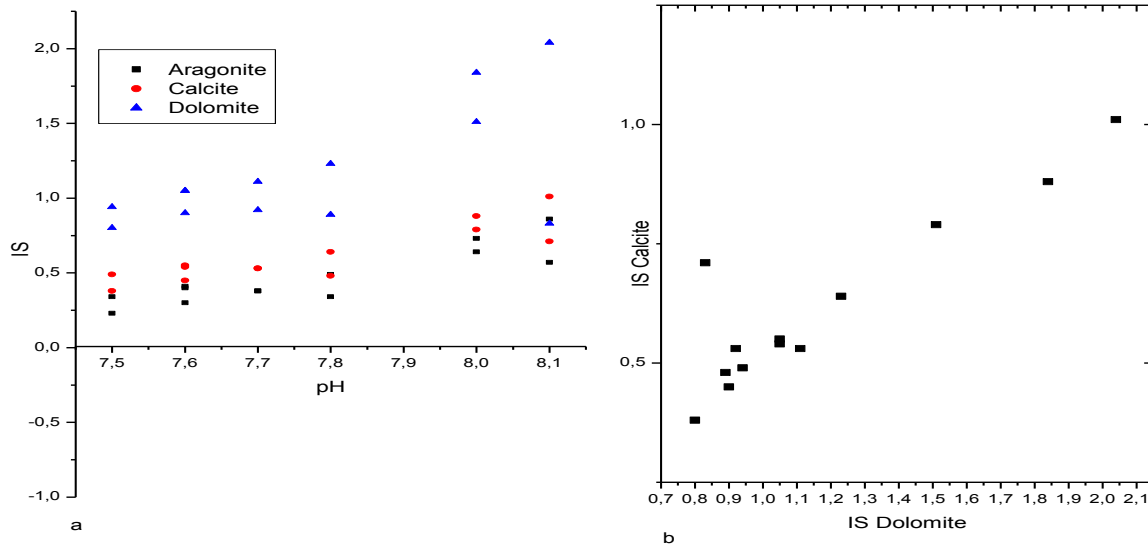


Fig. 9. Variation in the stability index of carbonate minerals by pH.

Pollution: Pollution is the consequence of the introduction of materials in quantities large enough to disrupt its normal functioning at short, medium or long term. Most of the time it is due to the activity of the man but not always (René, 2005). To properly understand the concept of effluent pollution, several factors that

characterize wastewater and the site where it is discharged must be incorporated. Several methods are adopted to determine the rate of this pollution in our region of study, we mainly talk about the organic pollution index (IPO), the microbiological quality index (IQM) and the biodegradability index (IB).

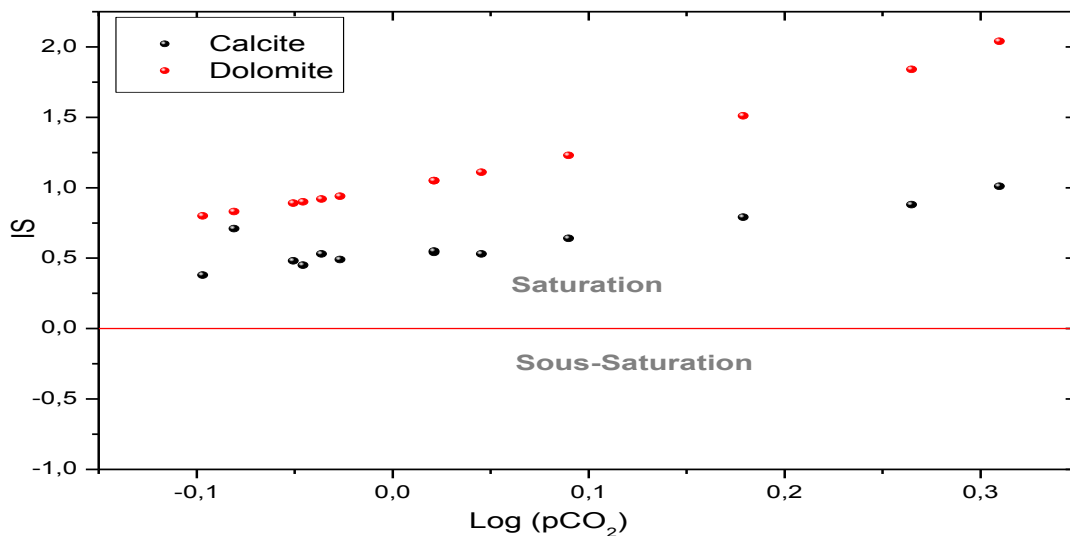


Fig. 10. Relation Log (Pco₂) vs IS Calcite and IS Dolomite applied to surface water.

Organic pollution index (IPO)

The detection of organic pollution in the surface waters of the study area is based on the organic pollution index (IPO). According to Leclercq and Maquet (1987), the principle of this method is to divide the pollutant element values into 5 classes, then to determine from its own measurements, the

corresponding class number for each parameter and then to average it (Table 3). There are two types of organic pollution, a low pollution for half of the samples (Fig. 11), the majority are taken upstream and a moderate organic pollution for samples taken downstream with the exception of the sample 10.

Table 3. Pollution Classification.

OPI		MQI	
Index	Class	Index	Class
5 - 4,6	Nullorganic pollution	4,3-5,0	Null
4,5 - 4	Loworganic pollution	3,5-4,2	Low
3,9 - 3	Moderateorganic pollution	2,7-3,4	Moderate
2,9 - 2	Strongorganic pollution	1,9-2,6	Strong
1,9 - 1	Verystrongorganic pollution	1,0-1,8	Verystrong

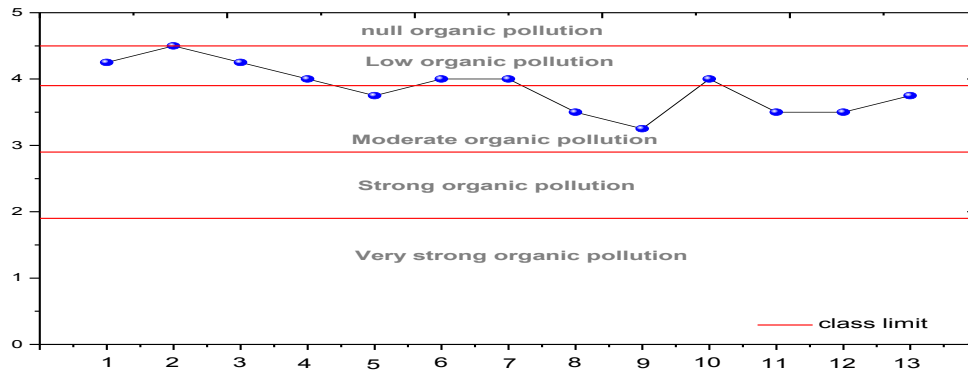


Fig. 11. Organic Pollution Index of Surface Water.

Biodegradability index (IB)

The self-purification or biodegradability classes are given by the biodegradability index (IB). These classes describe the state of the environment rich or poor in organic matter. It is an indicator of the potential for self-purification of water (Rassam *et al.*, 2012). If this ratio is less than 2, the effluent is easily biodegradable, when this ratio is between 2 and 3, the effluent is biodegradable with selected strains and when this ratio is greater than 3, the effluent is difficult to biodegrade. Surface water taken from upstream to downstream shows that the effluent is difficult to biodegrade (Fig. 12).

Microbiological quality index (IQM)

The presence of coliforms and streptococci testifies to a fecal contamination of these waters which it is imperative to purify them to preserve the natural environment (Olivier and Christelle, 1997). As for chemical analyzes, it is possible to calculate for rivers a bacteriological contamination index based on the enumeration of different germs, the main ones, generally associated with organic pollution, are total bacteria, fecal coliforms bacteria and fecal streptococci (Bovesse and Depelchin, 1980). The two samples taken, one upstream and the other downstream, make it possible to assess the evolution and the state of the microbiological pollution of the waters of the wadi Saf-Saf (Table 4).

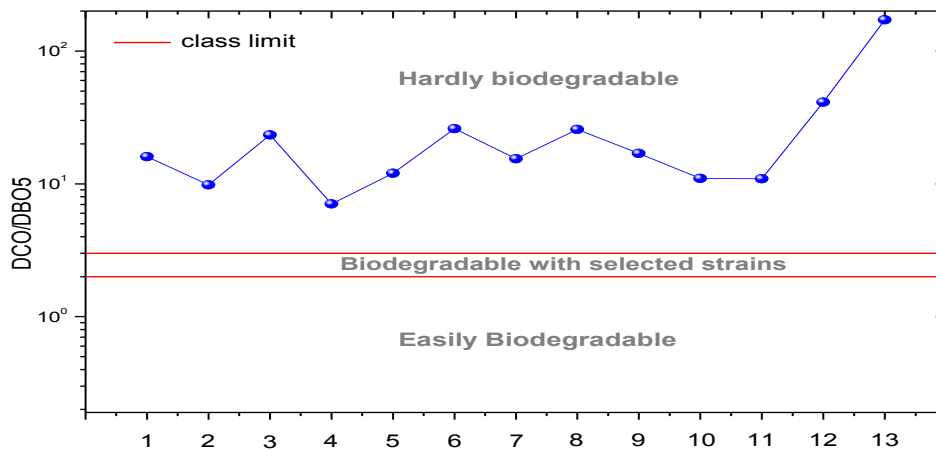


Fig. 12. Biodegradability index of surface water.

Table 4. Bacteriological analysis of surface water.

Parameters	Sample 1 (Upstream)	Sample 2 (Downstream)
Germes totaux à 22°C (UFC/ml)	150	160
Germes totaux à 37°C (UFC/ml)	200	180
Coliformes totaux à 37°C NPP/ml	24000	24000
Coliformes fécaux à 44°C NPP/ml	12000	12000
Stréptocoque fécaux à 37°C NPP/ml	Abs	Abs
Clostridium sulfito-réducteur à 46°C NPP/20ml	Abs	Abs

The bacteriological analyzes show a large number of fecal coliforms (12000 NPP/ml) and total (24000 NPP/ml). On the other hand, fecal streptococci and clostridium sulphito-reducers are absent in the two samples analyzed. The water analyzes show a microbiological quality index equal to 3, which gives a moderate fecal contamination.

Conclusion

The hydrogeochemical evolution of surface water concentrations described by the saturation index of the minerals in relation to the concentration factor shows a faster evolution of carbonates and the high sensitivity of these minerals to precipitation. The concentration of the carbonate alkalinity in the waters of the region can come from the dissolution of limestones or dolomites present both in the study area. A perfect evolution of NH_4^+ ammonium salts, ammonia NH_3^+ and phosphate ion PO_4^{3-} with the concentration factor, except for some samples. The high concentration of PO_4^{3-} is due to the supersaturation in hydroxyapatite and the high concentration of NH_3^+ and the perfect evolution of CO_2 , lead to poisoned surface water. The presence of lead speciation even at low concentrations indicates water toxicity which threatens aquatic life. Surface water quality has reached a threatening level. We have low to moderate organic pollution with effluents that are difficult to biodegrade and have a moderate fecal contamination.

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