African Journal of Environmental Science and Technology Vol. 4(8), pp. 526-534, August 2010
Available online at http://www.academicjournals.org/AJEST
ISSN 1991-637X © 2010 Academic Journals

Full Length Research Paper

Multivariate statistical characterization of groundwater quality in Ain Azel plain, Algeria

Lazhar Belkhiri[1*], Abdurrahman Boudoukha[2], Lotfi Mouni[3] and Toufik Baouz[4]

[1]Department of hydraulics, University of Hadj Lakhdar Batna, Batna 05000, Algeria.
[2]Laboratoire de recherche en hydraulique appliquée Université de Hadj Lakhdar Batna, Batna 05000, Algeria.
[3]Laboratoire de technologie des matériaux et de génie des procédés de l'université de Bejaia,
Targa-Ouzemour 06000, Alegria.
[4]Laboratory of Organic Materials, University of Bejaia, Targa- Ouzemour 06000, Algeria.

Accepted 25 May, 2010

Multivariate statistical techniques, cluster and principal component analysis were applied to the data on groundwater quality of Ain Azel plain (Algeria), to extract principal factors corresponding to the different sources of variation in the hydrochemistry, with the objective of defining the main controls on the hydrochemistry at the plain scale. Q-mode hierarchical cluster analysis grouped 54 groundwater samples into three clusters, that is, relatively less saline water (group 1), mixed water (group 2) and blended water (group 3), based on the similarity of groundwater quality characteristics. Principal component analysis, applied to the data sets of the three different groups obtained from hierarchical cluster analysis, resulted in four, five and three latent factors explaining 83.21, 83.36 and 87.30% of the total variance in groundwater quality data sets of group 1, group 2 and group 3, respectively. The varifactors obtained from PCA indicate that the parameters responsible for groundwater quality variations are mainly related to presence and dissolution of some carbonate, dolomitic and evaporite minerals; natural processes and water-rock interaction in the three water types. The results of this study clearly demonstrate the usefulness of multivariate statistical analysis in hydro chemical.

Key words: Cluster analysis, principal component analysis, hydrochemistry, Ain Azel plain, Algeria.

INTRODUCTION

In the current world economic paradigms, sustainable socioeconomic development of every community depends much on the sustainability of the available water resources. Water of adequate quantity and quality is required to meet growing household, industrial and agricultural needs. Groundwater quality is a very sensitive issue, which transcends national boundaries. It is influenced by many factors, including atmospheric chemistry, the underlying geology, the vegetation (or organic matter decay) and anthropogenic agents. The solubility of minerals in water places an upper limit on the maximum amounts of certain species of chemicals in natural waters. Some minerals like carbonates and evaporites dissolve quickly and change the composition of water faster, while other minerals like silicates dissolve more slowly and have less conspicuous effects on the composition of water. Temperature also plays a vital role in controlling the chemical and biological composition of a freshwater body. Previous studies (Frape et al., 1984; Garrels and McKenzie, 1967; Hem, 1989; Hartman et al., 2005) have revealed that the chemistry of natural waters can often be traced to the reaction of these waters with sediments or rocks through which they flow. Based on catchments studies in the USA, Walling (1980) observed differences in the weathering mechanisms of different rocks. Walling (1980) concluded that total dissolved solids in the water from limestones, volcanics and sand and gravel is almost independent of the amount of runoff. These differences in behaviour accrue from the differences in the solubilities of the minerals present in these rocks (Yidana et al., 2008).

Multivariate statistical techniques, cluster analysis (CA)

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*Corresponding author. E-mail: belkhiri_laz@yahoo.fr.

Abbreviations: CA, Cluster analysis; **FA,** factor analysis; **EC,** electrical conductivity; **PCA,** principal component analysis.

and factor analysis (FA), are effective means of manipulating, interpreting and representing data concerning groundwater pollutants and geochemistry. They are frequently employed to characterize the quality of groundwater. Liu et al. (2003) adopted FA to evaluate groundwater quality in a black foot disease area, Taiwan. Two factors, seawater salinization and arsenic pollutant, were considered and their factor scores were mapped. Love et al. (2004) applied FA to distinguish several groundwater signatures, including uncontaminated groundwater, agricultural activities, mining activities and sewage pollution. Olmez et al. (1994); Mahknecht et al. (2003); Farnham et al. (2003) used FA to discuss geochemical evolution and mineralization and groundwater contamination. Additionally, the CA was also used to interpret the hydrochemical data based on factor scores, for example, Suk and Lee (1999); Reghunath et al. (2002); Kim et al. (2005). This study comprises application of multivariate statistical techniques to groundwater quality data set obtained from groundwater of Ain Azel plain in Algeria.

DESCRIPTION OF THE STUDIED AREA

The studied area is located in the east of Algeria. Most of inhabitants (more than 30000 inhabitants) are centered on the town of Ain Azel. The principal activity in this area is the production of cereals (barley and corn) and its climate is semi-arid. The amount of rainfall varies from place to place and from season to season, ranging from 346 mm/yr in the plains (altitude < 1000 m) to 534 mm/yr in the mountains (altitude > 1000 m). The annual maximum and minimum temperatures are 21.8 and 8.5°C, respectively (Belkhiri, 2005).

According to many authors (Savornin, 1920; Galcon, 1967; Guiraud, 1973; Vila, 1980) the area of concern is distinguished by two geological sets. In the South, autochthonous Jurassic and Cretaceous carbonate lithologies are mainly observed in Djebels Boutaleb, Djebel Hadjar Labiod and Fourhal (Figure 1). In the North, allochthonous Jurassic and Cretaceous lithologies are observed in Djebels Kalaoun and Sekrine. A lithostratigraphic study of Ain Azel area identifies the following two aquifer formations. The first is a carbonate and sandy formation of about 700 m thick presenting fracture porosity and constitutes the Barremian formation. The second is an alluvial formation of about 250 m thick corresponding to a Mio-Plio-Quaternary formation with interstitial porosity (Boutaleb, 2001; Belkhiri, 2005). A mineralogical study shows that the metalliferous minerals, particularly the sphalerite, the gangue minerals and the dolomite contain variable quantities of trace metals such as Zn, Pb, Cd, Fe and Cu (Boutaleb, 2001).

The studied area is situated in the alluvial plain of the Mio-Plio-Quaternary (Figure 1) showing a heterogeneous continental detrital sedimentation (Boutaleb, 2001; Belkhiri, 2005; Attouche, 2006). This aquifer is directly fed by stream water coming from different relief's surrounding the depression inter-mountainous of Ain Azel. The plain hosts a large number of water-wells with depths varying from 8 - 38 m. Most of these wells supply water for drinking and irrigation. The direction of groundwater flow around Ain Azel plain is from south to north and from west to east. In general, the groundwater flows toward center of the plain (Figure 2) and development this map shows a piezometric depression, which coincides with high density of the well in the center of the plain. The limits of south and west are of imposed entering flux; however the limit of east is of null flux. The pumping tests on different wells showed high transmissivity (30 - 36 m^2/day) indicating high yields (Belkhiri, 2005).

MATERIALS AND METHODS

Sample collection and analysis

18 wells currently in use were selected based on the preliminary field survey carried out to understand the overall distribution of the various types of wells in the studied area (Figure 1). The selected wells are used for domestic, agricultural, and domestic/agricultural purposes and are uniformly distributed over the area of concern. Groundwater samplings were performed three times in 2004: June, September and December and a total of 54 groundwater samples were collected during this period. The samples were collected after pumping for 10 min. This was done to remove groundwater stored in the well. These samples were collected using 4 - 1 acid-washed polypropylene containers. Each sample was immediately filtered on site through 0.45 µm filters on acetate cellulose. Filtrate for metals analyses were transferred into 100-cm^3 polyethylene bottles and immediately acidified to pH < 2 by the addition of Merck^{TM} ultrapure nitric acid (5 ml 6 N HNO3). Samples for anions analyses were collected into 250-cm^3 polyethylene bottles without preservation. All the samples were stored in an ice chest at a temperature of < 4°C and later, transferred to the laboratory and stored in a refrigerator at a temperature of < 4°C until analyzed (within 1 week). Immediately after sampling, pH and electrical conductivity (EC) were measured in the field using a multi-parameter WTW (P3 MultiLine pH/LF-SET).

Subsequently, the samples were analyzed in the laboratory for their chemical constituents such as calcium, magnesium, sodium, potassium, chloride, bicarbonate, sulfate, lead and iron. This was achieved using standard methods as suggested by the American Public Health Association (APHA, 1989; 1995). Ca, Mg, HCO_3 and Cl were analyzed by volumetric titrations. Concentrations of Ca and Mg were estimated titrimetrically using 0.05 N EDTA and 0.01 N and those of HCO_3 and Cl by H_2SO_4 and $AgNO_3$ titration, respectively. Concentrations of Na and K were measured using a flame photometer (Model: Systronics Flame Photometer 128) and that of sulfate by turbidimetric method (Clesceri et al., 1998). Standard solutions for the above analysis were prepared from the respective salts of analytical reagent grades. Trace metals were determined by Graphite Furnace Atomic Absorption Spectrophotometer (Perkin-Elmer AAnalyst 700) using multi element Perkin-Elmer standard solutions. The accuracy of the chemical analysis was verified by calculating ion-balance errors where the errors were generally around 10%.

Multivariate statistical analysis

In recent times, multivariate statistical methods have been applied

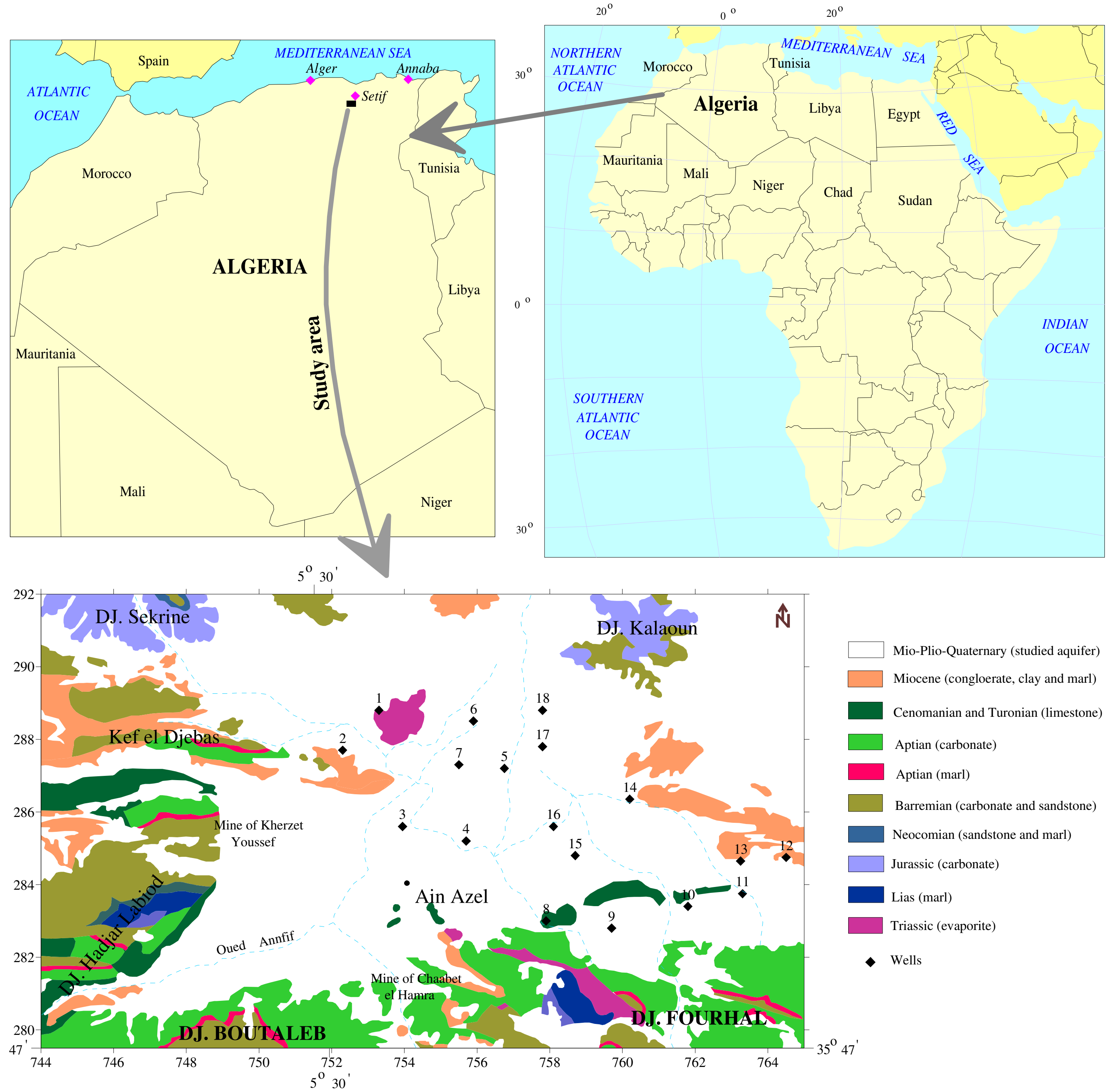

Figure 1. Map showing water sampling locations and geology of the studied area.

widely to investigate environmental phenomena e.g., (Anazawa et al., 2003; Anazawa and Ohmori, 2005; Güler and Thyne, 2004; Laaksoharju et al., 1999). The combined use of principal component analysis (PCA) and cluster analysis enabled the classification of water samples into distinct groups on the basis of their hydrochemical characteristics. Multivariate statistical tools have been successfully used to study and classify different sediment types (Huisman and Kiden, 1998; Tebens et al., 2001), and hydrogeochemical processes (Cameron, 1996; Duffy and Brandes, 2001; Gupta and Subramanian, 1998). Momen et al. (1996) used cluster analysis and PCA to identify the temporal and spatial variation of water chemistry in Lake George in New York. Tariq et al. (2005) similarly applied multivariate techniques to trace metal levels in tannery effluents in Pershawar in Pakistan. Their study involved samples from tannery effluents, groundwater and soils and with the aid of multivariate tools they were able to

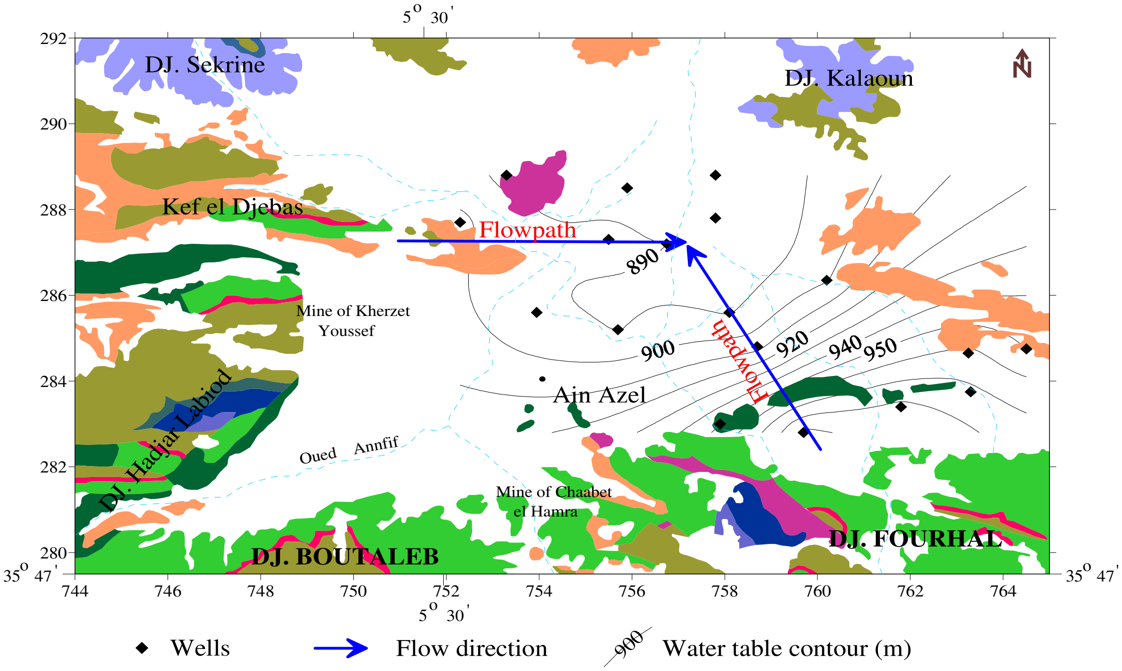

Figure 2. Potentiometer surface map in December, 2004 (unit: m).

correlate important chemical species of the three media and established significant relationships. Using factor analysis, Zeng and Rasmussen (2005) attributed the variations in quality of water from Lake Lanier in Georgia to anoxia associated with lake stratification. This paper dwells on the strength of multivariate techniques to characterize the hydrochemical variations along the Ain Azel plain. It employs the combined use of cluster and factor analysis to assess the spatial and temporal variations of groundwater chemistry.

Cluster analysis

Cluster analysis was used to determine if the samples can be grouped into statistically distinct hydrochemical groups that may be significant in the geologic context. A number of studies used this technique to successfully classify water samples (Alther, 1979; Williams, 1982; Farnham et al., 2000; Alberto et al., 2001; Meng and Maynard, 2001). Comparisons based on multiple parameters from different samples were made and the samples were grouped according to their 'similarity' to each other. Classifications of samples according to their parameters are known as Q-mode classifications. In the present study Q-mode HCA was used to classify the samples into distinct hydrochemical groups. The Ward's linkage method (Ward, 1963) was used in this analysis. A classification scheme using Euclidean distance (straight line distance between two points in *c*-dimensional space defined by *c* variables) for similarity measurement, together with Ward's method for linkage, produces the most distinctive groups where each member within the group is more similar to its fellow members than to any member outside the group (Güler et al., 2002). All 11 hydrochemical variables measured (consisting of EC, pH, Ca, Mg, Na, K, Cl, SO_4, HCO_3, Pb and Fe) were utilized in this analysis. For statistical analysis, all the variables were log-transformed and more closely correspond to normally distributed data. Subsequently, they were standardized to their standard scores (z-scores) as described by Güler et al. (2002).

Hydrochemical results of all samples were statistically analyzed by using the software STATISTICA®.

Factor analysis

Factor analysis is a multivariate analytical technique, which derives a subset of uncorrelated variables called factors that explain the variance observed in the original dataset (Anazawa and Ohmori, 2005; Brown, 1998). Factor analysis is used to uncover the latent structure of a set of variables. In technical terms, common factor analysis represents the common variance of variables, excluding unique variance and is thus a correlation-focused approach seeking to reproduce the intercorrelation among the variables. On the other hand, components (from PCA) reflect both common and unique variance of the variables and may be seen as a variance-focused approach that reproduces both the total variable variance with all components as well as the correlations. PCA is far more commonly used than principal factor analysis (PFA). However, it is common to use "factors" interchangeably with "components" in multivariate analysis. Factor analysis can be performed on any kind of scientific data to establish a pattern of variation among variables or reduce large data sets into factors for easy handling and interpretation.

The total number of factors generated from a typical factor analysis indicates the total number of possible sources of variation in the data. Factors are ranked in order of merit. The first factor or component has the highest eigenvector sum and represents the most important source of variation in the data. The last factor is the least important process contributing to the chemical variation. Factor loadings on the factor loadings tables are interpreted as correlation coefficients between the variables and the factors. In this research, PCA was applied to chemical data from the Ain Azel plain to extract the principal factors corresponding to the different sources of variation in the data. Here, PCA was selected for the reasons stated above. In order to maximize the variation among the variables under each factor, the factor axes were subsequently varimax rotated.

Table 1. Statistical summary of hydrochemical parameters of groundwater.

		EC	pH	Ca	Mg	Na	K	Cl	SO_4	HCO_3	Pb	Fe
June (n = 18)	Min	545	6.7	64.13	35.88	18.5	6.14	56.80	14	131.76	0.03	0.023
	Max	1783	7.9	232.46	181.20	143.7	18.02	337.25	309	1348.10	1.83	0.338
	Mean	1012	7.1	134.13	84.20	61.7	12.92	166.66	114	392.70	0.61	0.149
	Std.Dev.	374	0.3	45.76	41.54	37.9	3.20	94.82	82	256.52	0.53	0.082
Sept (n = 18)	Min	470	6.8	64.12	35.88	16.7	5.14	56.80	18	176.90	0.29	0.067
	Max	2203	7.6	256.51	184.20	145.3	19.01	330.15	312	1342.00	1.79	0.789
	Mean	1092	7.2	132.84	87.42	61.3	12.11	160.18	118	421.58	0.91	0.410
	Std.Dev.	533	0.2	46.67	46.42	38.2	3.49	87.58	82	240.37	0.45	0.211
Dce (n = 18)	Min	629	6.8	62.52	35.45	16.3	4.20	63.90	37	183.00	0.02	0.089
	Max	2578	7.9	250.09	192.21	145.4	20.20	337.25	368	1335.90	0.29	0.645
	Mean	1304	7.1	128.49	83.96	60.3	12.04	170.99	134	433.32	0.09	0.325
	Std.Dev.	604	0.3	45.53	39.96	38.3	3.47	94.91	90	242.17	0.07	0.133
Total (n = 54)	Min	470	6.7	62.52	35.45	16.3	4.20	56.80	14	131.76	0.02	0.023
	Max	2578	7.9	256.51	192.21	145.4	20.20	337.25	368	1348.10	1.83	0.789
	Mean	1136	7.1	131.82	85.19	61.1	12.36	165.94	122	415.87	0.54	0.295
	Std.Dev.	518	0.3	45.18	41.94	37.4	3.35	90.85	84	242.38	0.53	0.185

All values are in mg/l except pH and EC (µ.Siemens/cm).

RESULTS AND DISCUSSION

Hydrochemical characteristics

Statistics of the chemical compositions of the groundwater samples are shown in Table 1. The groundwater samples of the study area have pH values ranging from 6.7 - 7.9, which indicate that the groundwater is slightly alkaline. The electrical conductivity (EC) values ranged 470 - 2,578 µS/cm. The order of abundance of the major cations is Ca ≥ Mg > Na > K and all samples exceeded the desirable limit of Ca for drinking water (75 mg/l) except sample 10, but only 78% of them exceed that of Mg (50 mg/l). The abundance of the major anions is $HCO_3 \geq Cl > SO_4$ and almost 28% of the samples exceeded the desirable limit of Cl (200 mg/l), but the sulfate concentrations are all below health guidelines (WHO, 1993). The concentration of lead for the three campaigns ranges from 0.02 - 1.83 mg/l and all samples exceeded the WHO guideline limit of 0.01 mg/l. In the case of iron, the concentration in many of the samples was higher than the WHO permitted limit of 0.3 mg/l and the percent samples above the WHO limit ranges from 60 - 83% for the three campaigns. Since in this region, except for agricultural activity no other anthropogenic activities were being carried out in this region and hence the high Fe concentration in these waters could not be assigned to the anthropogenic activities but rather the water-rock interaction should be the reason for the higher value of Fe observed in the plain. It is clearly observed that the concentrations of the major elements do not undergo an appreciable change during the three campaigns. For this reason the three campaigns were grouped as one.

Cluster analysis

Cluster analysis suggests three groups of groundwater's (Figure 3 and Table 2). EC seems to be a major distinguishing factor, which increases with concentrations increasing in all major-ions following the order: Group 1, 2 and 3 (Table 2). The Group 1 is composed of the wells 3, 4, 5, 6, 7, 10, 12 and 13, and concerns 44% of the water samples. This type of water is relatively fresh with a mean EC of 782 µS/cm, which is the characteristic of less saline water (Ca-Mg-HCO_3). This group is basically bicarbonate dominated, however; calcium and magnesium are also present.

Group 2 is represented by the wells 1, 8, 11, 15, 16 and 17, and it occupies 24% of the water samples. The electrical conductivity for this group is 1.062 µS/cm, which is the characteristic of mixed water (Mg-Ca-HCO_3-Cl). This water type 2 is bicarbonate and chloride dominated and it also has low concentrations of sulfate. This water type can be interpreted as the first step of water-rock interactions occurring in dilute solutions, explained by the abundance of carbonate and evaporite formations in the aquifer.

Group 3 includes samples: 2, 9, 14 and 18, where the EC is 1.956 µS/cm which is the characteristic of blended water (Mg-Ca-Cl-HCO_3). Chloride content is also high with respect to bicarbonate concentration. Based ongeological

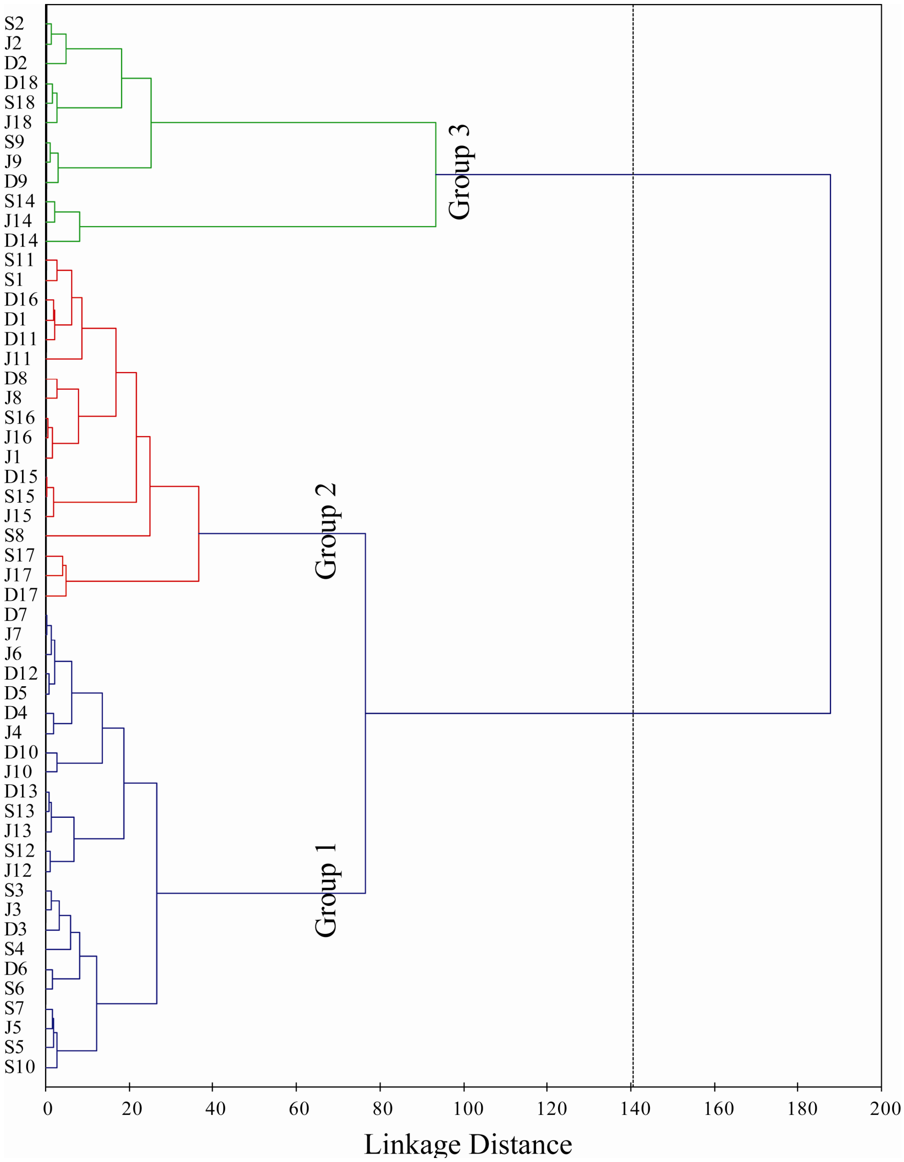

Figure 3. Dendrogram of the hydrochemical samples.

Table 2. Mean parameter values of the three principal water groups (determined from HCA).

Group	EC	pH	Ca	Mg	Na	K	Cl	SO_4	HCO_3	Pb	Fe
G1(n = 24)	782	7.0	108.93	54.98	26.3	10.81	82.83	98	352.03	0.45	0.319
G2 (n = 18)	1062	7.1	120.08	89.44	78.0	12.37	198.05	127	342.62	0.54	0.336
G3(n = 12)	1956	7.3	195.22	139.25	105.4	15.43	284.01	164	653.41	0.70	0.185

pH (standard units), EC (µ.Siemens/cm) and mean (mg/l).

and geographical position, discharges of industrial wastewater and contamination of the groundwater by seawater were excluded as the source of high concentrations. A local contamination by septic tanks, sewage systems, agricultural fertilizers and dissolution of the formations clay, marl and salt and could be responsible for this elevation.

Factor analysis

The Kaiser criterion (Kaiser, 1960) was applied to

Table 3. Rotation PCA loading matrix

Group	**1**				**2**					**3**		
Component	**1**	**2**	**3**	**4**	**1**	**2**	**3**	**4**	**5**	**1**	**2**	**3**
EC	0.92	0.22	-0.18	0.06	0.20	-0.09	-0.02	0.92	-0.14	-0.13	0.69	0.62
pH	-0.16	-0.28	0.37	0.78	0.80	0.21	-0.22	0.14	0.07	-0.96	-0.07	-0.07
Ca	0.81	0.05	0.02	-0.27	-0.86	0.25	0.01	0.11	0.21	0.44	0.84	-0.15
Mg	0.81	0.26	0.12	-0.28	0.13	0.87	0.20	0.05	0.03	0.68	0.62	0.10
Na	0.34	-0.82	0.06	0.26	-0.28	0.37	-0.59	0.37	0.43	0.93	0.29	-0.07
K	0.23	-0.26	-0.46	0.63	0.05	-0.14	0.07	-0.14	0.93	0.24	-0.93	-0.04
Cl	0.88	-0.38	-0.07	0.06	0.78	0.27	0.07	0.36	0.25	0.93	0.18	-0.20
SO_4	-0.47	0.06	-0.15	0.77	0.05	0.14	0.94	-0.04	0.07	0.91	-0.35	-0.06
HCO_3	0.40	0.78	0.00	-0.36	-0.40	0.07	0.84	0.09	0.06	-0.91	0.05	0.32
Pb	-0.01	0.09	0.89	0.00	0.35	0.74	-0.02	-0.35	0.05	-0.66	0.05	-0.23
Fe	0.23	0.78	0.36	0.13	-0.16	0.72	-0.04	0.05	-0.16	0.00	-0.02	0.91
% of variance	32.32	20.86	12.24	17.80	22.13	19.93	18.47	11.80	11.02	49.72	24.45	13.14
Cumulative %	32.32	53.17	65.41	83.21	22.13	42.06	60.54	72.34	83.36	49.72	74.17	87.30

The bold values indicate absolute component loadings higher than 0.5, which are considered significant contributors to the variance in the hydrochemistry.

determine the total number of factors for each data set in this analysis. Under this criterion, only factors with eigenvalues greater than or equal to 1 will be accepted as possible sources of variance in the data, with the highest priority ascribed to the factor that has the highest eigenvector sum. The rationale for choosing 1 is that a factor must have a variance at least as large as that of a single standardized original variable to be acceptable.

Group 1

Four principal components (PC) were extracted and rotated using the varimax normalization (Kaiser, 1960). An initial run using the Kaiser criterion (Kaiser, 1960) resulted in five principal components. However, it was observed that the fifth factor would not constitute a unique source of variance in the hydrochemistry since it had only one loading greater than 0.50. It was therefore dropped and four factors were chosen for varimax rotation. The results of (Table 3) show that the four PC account for more than 83.21% of the total variance, which is quite good and can be relied upon to identify the main sources of variation in the hydrochemistry. PC1 represents about 32.32% of the variance and has high absolute loadings for EC, Ca, Mg and Cl and probably shows the result of mineral water reactions in the area. PC 2, which accounts for 20.86% of the total variance, contains high loadings for Na, HCO_3 and Fe. PC1and PC2 represents the major geochemical processes taking place, which is most probably the presence and dissolution of some carbonate, dolomitic and evaporite minerals in the aquifer. PC3 and PC4, respectively, show high loadings for Pb and pH, K and SO_4. PC3 and PC4, respectively, account for 12.24 and 17.80% of the variation in the hydrochemistry.

Group 2

Five significant PCs explain 83.36% of the total variation in the hydrochemistry. Most of the variance is contained in the PC1 (22.13%), which is associated with the variables pH, Ca and Cl (Table 3). PC 2 represents 19.93% of the total variation in the hydrochemistry and has high loadings for Mg, Pb and Fe. The variables Na, HCO_3 and SO_4 contribute most strongly to the third component (PC3) that explains 18.47% of the total variance. PC1, PC2 and PC3 are assumed to be indicative of the natural processes and water-rock interaction. PC4 and PC5, respectively, show high loadings for EC and K.

Group 3

Three PCs explain 87.30% of the total variance (Table 3). Most of the variance is contained in the PC1 (49.72%), which is associated with the variables pH, Mg, Na, Cl, SO_4, HCO_3 and Pb and probably shows the result of mineral water reactions in the plain. The aquifer is sedimentary rocks, which range from sandstones to conglomerates to clays with varying mineralogical compositions. The variables EC, Ca and K contribute most strongly to the third component (PC2), which explains 24.45% of the total variance and probably represents the presence of carbonate minerals in the aquifer. PC3 explains 13.14% of the variance and is mainly related to Fe and assumed to be indicative of the water-rock interaction.

CONCLUSION

Multivariate statistical techniques including cluster and principal component analysis can successfully be used to derive information from the data set about the possible influences of the environment on groundwater quality and also identify natural groupings in the set of data. These methods are important to avoid misinterpretation of environmental monitoring data due to uncertainties. In this study, multivariate statistical methods were applied to data set obtained from Ain Azel plain, Algeria. Interpretation of analytical data showed that the abundance of the major ions is as follows: $Ca \geq Mg > Na > K$ and $HCO_3 \geq Cl > SO_4$. Three major water types are suggested by the Q-mode HCA analysis. The samples from the area were classified as less saline water (Ca-Mg-HCO_3), mixed water (Mg-Ca-HCO_3-Cl) and blended water (Mg-Ca-Cl-HCO_3). The varifactors obtained from PCA indicate that the parameters responsible for groundwater quality variations are mainly related to presence and dissolution of some carbonate, dolomitic and evaporite minerals in relatively less saline water type; natural processes and water-rock interaction in mixed water type; water-rock interaction in blended water type. The results of this study clearly demonstrate the usefulness of multivariate statistical analysis in hydro chemical.

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