

CHEMICAL CHARACTERIZATION AND STATISTICAL ANALYSIS OF BOTTLED WATERS IN ALGERIA

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ABSTRACT

Fifty-six brands of bottled water marketed on the Algerian market are the subject of this study. Hydrochemical classification and statistical distribution based on multivariate analysis using appropriate tools were used to analyze data reported on bottled water labels. These waters were together into groups with similarities in terms of chemical characteristics and mineral content. The results obtained offer consumers a simple tool for assessing the differences between bottled waters, as to the choice of water to acquire.

KEY WORDS: Algeria, Bottled water, Ionic balance, Piper diagram, Statistical analysis

INTRODUCTION

Water is a vital constituent, but its quality has been seriously degrading for many years under the influence of various pollution factors, chemical, organic and microbiological. These factors are mainly due to the overcrowding of the population leading to the diversification of human economic activities (Ramdani 2012; Merouche, 2019).

In arid and semi-arid regions, which is characterized by high rainfall irregularity such as Algeria, domestic, industrial and irrigation water needs have increased considerably in recent decades (Kadi, 1997; Merouche, 2019). However, socio-economic development and rapid urbanization have had a negative impact on the quality of water resources. Many cases of industrial and urban pollution have been observed in this case at the level of dams, groundwater and rivers over the past two decades (Kadi, 1997; Baba, 2007).

However, tap water is drinkable, but its quality is not guaranteed, due to problems of infiltration in the pipes in the event of rain, for example (Saleh, 2001; Blackburn *et al.*, 2004; Mouhoumed *et al.*, 2020) or the presence of certain organic compounds, which cannot be completely removed during conventional treatment steps (Zhan *et al.*, 2010). Such examples

convince the public to use bottled mineral water (Baba, 2007). The reasons for this choice are mainly aesthetic and closely related to unpleasant tastes and odors (Saleh, 2001; Mouhoumed *et al.*, 2020; Hadbi and Hamoudi Abdeamir, 2020).

Just 20 years ago, water in a plastic bottle was not a recurring item on many consumers' shopping lists. Today, billions of liters of water are sold in all types of packaging and containers (Al Rayes *et al.*, 2013). Bottled water represents one of the fastest growing beverage markets in the world (Versari, 2002; Samek 2004) and recent projections indicate that it is likely to overtake non-alcoholic beverages and become the most large beverage category by volume (Mouhoumed *et al.*, 2020).

Alongside the global market, the bottled water market in Algeria has not always been as it is today, in full swing with a great diversity of bottled water supply. The first national mineral water company was created in 1966 to exploit, produce, bottle and market the oldest bottled water in Algeria "Saida" (Hazzab, 2011). For around twenty years, the bottled water market in Algeria has experienced strong growth; this development has materialized through the establishment of several operating and production units throughout the national territory (Hazzab, 2011).

Currently there are more than fifty brands between mineral waters and spring waters produced all over the country with a concentration in the center, particularly in the wilaya of Bejaia.

Natural mineral waters are characterized by their original purity and by the stability of their composition in minerals and trace elements, likely to give them certain therapeutic virtues according to health specialists (Petraccia, 2006; Baba, 2007; Degrémont, 1989; Astel, 2014; Hegui, 2020). These specialists specify that each water has preferential indications depending on its composition and mineral content (Mice, 2008; Astel, 2014).

This article abstracts a simple classification that makes it possible to differentiate bottled waters in Algeria according to their composition, in order to guide citizens who are more and more inclined to drink mineral and spring waters.

The results obtained can constitute a tool to help and assess the decision as to the choice of water to acquire, and this for an informed consumer as well as for a nutrition specialist. It is expected that this information will also be useful to growers and regulatory bodies.

Regulatory framework for the exploitation of mineral water in Algeria

Within the meaning of the legislative texts adopted in Algeria, only two types of water intended for packaging are defined: natural mineral water and spring water (Executive Decree n°04-196 of July 15, 2004 published in the Official Journal n°45 of July 18, 2004). Natural mineral water is microbiologically healthy water coming from an aquifer or an underground deposit, exploited from one or more natural or drilled emergences, near which it is packaged. Natural mineral water has a stable composition over time, and is clearly distinguished from other water intended for human consumption by its purity, and its specific content of mineral salts, trace elements or other constituents.

Spring water is water of exclusively underground origin, suitable for human consumption, microbiologically healthy and protected against the risk of pollution. It must be introduced to the place of its emergence, as it comes out of the ground. It is less mineralized and its composition does not have the same degree of stability and can vary while remaining within the limits respecting the potability standards in force.

These two types of water cannot be subject to any specific treatment or addition. Their quality

characteristics are defined by (Interministerialdecree of January 22, 2006 published in the Official Journal n° 27 of April 26, 2006) and (Interministerialdecree of October 23, 2014 published in the Official Journal n° 3 of January 27, 2015).

MATERIALS AND METHODS

Bottled Water Database

Fifty-six brands of water sold in bottles on the Algerian market were used for this study. They were collected from local supermarkets and food stores, between July and September 2021.

The data, relating to the chemical characteristics of the main brands of water, were recorded from the labeling of the various products, stuck on the bottles (Tables 1 and 2). They correspond to the results of analyzes carried out in laboratories officially authorized to carry out the water characterization operation, in accordance with the regulations in force in Algeria.

However, it is interesting to control the quality and the consistency of the analysis carried out in major ions of bottled water. Nevertheless, a fundamental condition of electrolyte solutions is that at the molecular scale, a condition of electro-neutrality exists (equation 1) (Freeze and Cherry, 1979).

$$(\Sigma\text{cations}) = (\Sigma\text{anions}) \quad (1)$$

To can be able to present and interpret the results of analyses, it is common practice to express the deviation compared equality in the form of equation (2) where E is the load balance error expressed percentage (Freeze and Cherry, 1979; Appeloand Postma, 2005).

$$E (\%) = \frac{\Sigma\text{cations} - \Sigma\text{anions}}{\Sigma\text{cations} + \Sigma\text{anions}} \times 100 \quad (2)$$

This validation process is a prerequisite for a reliable presentation of the analysis results and their interpretation (Labadi and Hammache, 2016; Mouhoumed *et al.*, 2020).

Deviations from electro-neutrality arise from random or systematic errors in one or more constituents or from incomplete analysis that overlooks some significant constituents (Hem, 1985; Olive, 1996). Balance errors of up to 2% are unavoidable in almost all water analysis laboratories. Sometimes even greater error must be accepted, but with deviations greater than 5%, sampling and analysis procedures must be considered (Freeze and Cherry, 1979; Olive, 1996;

Table 1. Chemical composition indicated on the labels of mineral waters bottled in Algeria.

N°	Brand Name	pH	mg/l										
		pH Unit	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	HCO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	F ⁻	NO ₃ ⁻	NO ₂ ⁻	TDS
1	Batna	6.9	59	45	15	2	378.2	22	40		00	00	561,2
2	Chifaa	7.2	66	26	63	8	263	48	40		11		525
3	Daouia	7.8	31.9	19.9	75	0.7	288	40.4	18.3		0.4		474,6
4	Djemila	7.04	98	20	100	3	281	170	50		5	0	727
5	El Goléa	7.3	35	16	36	5	-	21	32		-	00	145
6	Guedila	7.35	78	37	29	2	317	40	95		4.5	<0.01	602,5
7	Hammamat	7.38	62.9	15.1	13.2	4.4	208.8	20.4	28.6		5.1	0.01	358,5
8	Ifri	7.2	99	24	15.8	2.1	265	72	68		15	<0.02	560,9
9	Lalla Khedidja	7.22	53	7	5.5	0.5	160	11	7	0.3	0.42	00	244,7
10	Manbaa	7.1	93	31	68	4	326	84	153	1.1	8.9	0.02	769
11	Mansourah	7	85	37	30	1	362	48	53		12	0	628
12	Messergline	7.2	52	42	45	3	260	78	50		5	00	535
13	Milok	7.56	59	12	9	3	152	16	65		15.2	0	331,2
14	Mouzaïa	6.5-7.5	136	75	145	3	600	150	85		8	0.02	1202
15	N'Gaous	7.66	143	65.4	63.4	3.8	-	75	44.4	1.2	2.07	00	398,3
16	Saïda	7.5	68	50	58	02	376	81	65		15	00	715
17	Sfid	7.12	74	35	28	2	268	68	91		<28	0	594
18	Sidi driss	7.69	38.9	3.2	7.1	0.7	127.2	17	10		0.2	0.02	204,3
19	Sidi Okba	7.06	143	65.4	63.4	3.8	213	75	445		2.1	00	1010,7
20	Sidi Yacoub	6.5	83	14	37	4.9	226	65	32		0	0	461,9
21	Texanna	7	30	9.1	11	1	60	28.4	11		00	00	150,5
22	Thevest	7.77	90	34.1	47.3	1	231.8	65	188		2.35	<0.01	659,6
23	Toudja	7.19	56.6	15.2	29	0.7	-	48	19.6		4	<0.01	173,1
24	Youkous	7.4	77.0	14.5	13.4	4.7	219	25.7	35.8		2	00	392,1

Appelo and Postma, 2005).

Chemical classification of bottled waters

Generally the typical facies of water analyzed is defined by determining the main anion and the main cation. But if we have many analyzes carried out in a region, it is in our interest to define a certain number of water families using Piper's trilinear diagram (Olive, 1996).

The Piper diagram makes it possible to present on the same diagram a large number of analyzes which can be compared visually in a simple way. It is composed of two triangles, representing the cationic facies and the anionic facies, and a diamond synthesizing the global facies. The point clouds concentrated in a pole represent for the different samples the combination of cationic and anionic elements (Piper, 1944; Freeze and Cherry, 1979; Olive, 1996).

Statistical analyzes of data

The application of statistical analysis on complex data sets has aroused great scientific interest. The waters studied underwent a statistical study based on a Hierarchical Cluster Analysis (HCA) and a

Principal Component Analysis (PCA) using the XLSTAT software package.

HCA allows large data to be classified into groups based on similarities or dissimilarities. Thus, the resulting groups are similar to each other but distinct from other groups. Euclidean distance was used as a measure of similarity. Ward's method was used as the partition method.

Bottled water brands have been classified according to their major ion composition. The result of the HCA is presented in the form of dendrograms with the number of groups selected based on a visual examination of the dendrogram.

In order to characterize the water groups thus identified, a Principal Component Analysis (PCA) followed by a graphical representation of the individuals in the first factorial planes was also carried out on the same data.

RESULTS AND DISCUSSION

Chemical classification

Ionic balance

Nine bottled mineral waters (Chifaa, El Goléa, Lalla

Table 2. Chemical composition indicated on the labels of spring waters bottled in Algeria.

N°	Brand Name	pH	mg/l										TDS
			Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	HCO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	F ⁻	NO ₃ ⁻	NO ₂ ⁻	
1	Ain Bouglez	6,87	4.6	3.8	29	1	-	30	10		9	0.06	87,5
2	Alma	7	91	37	31	2	350	55	87		15	<0.01	668
3	Ariaf	7.25	94.4	32.1	96.3	1.5	202.1	198	163.1		6.2	00	793,7
4	Arwa	7.33	120	23	56	1	256	100	104		46,5	<0.01	706,5
5	Ayris	7.78	65.6	6.8	28.5	1.9	234.2	37	75		2.7	0.01	451,7
6	Besbassa	7.29	54.2	2.6	5	2	164.7	10	4		9	<0.01	251,5
7	Bir Salam	7.44	62.9	13.5	22.8	2.1	-	23.1	71.5		<15	<0.02	210,9
8	Djebel Amour	7.13	81	14	21	0.2	198	38.4	64		29.3	<0.01	445,9
9	Djurdjura	7.67	103	28	54	1	357	97	56		30	<0.01	726
10	El Djazia	7.21	103	27	34	2	351	43	64	0.3	20.2	0	644,5
11	El Ghadir	7.5	111	28	25	3	317	37	106		25	<0.01	652
12	El kantara	7.32	90	37	36	3	247	59	162		9.6	<0.01	643,6
13	Fezguia	7.46	85.7	13.4	31	1	305	30	28		19	00	513,1
14	Guerioune	7.28	72	27	11	2	336	21	11		20.2	<0.01	500,2
15	Hirouche	7.20	107.4	18.2	22.2	1.6	262.8	62.2	54.0	0.2	33	<0.01	561,6
16	Ichemoul	7.52	101	2	10	0.6	262	10	54		13	<0.01	452,6
17	Ifren	7.5	68.8	10.7	32	2.4	283	17	62.5		3.2	<0.01	479,6
18	Ledjdar	7.53	64	37	30	4	308	41	66		<50	<0.1	600,1
19	Medjana	7.12	136	42	62	2	458	47	211	0.3	1.8	0.01	960,1
20	Messâd	7.13	79	27	50	2	275	40	156		2.3	<0.01	631,3
21	Mileza	7.33	111	34	29	1	311	10	190		3.2	<0.01	689,2
22	Nestlé	7.8	55	17	>12	0.5	210	>15	33		4.6	00	347,1
23	Ouwis	7.42	106	25	60	2	261	48.6	177		18.3	<0.01	697,9
24	OVitale.	6.92	91	14	30	1	214	50	86		<15	0	501
25	Qniaa	7.24	111.7	27	48.2	2.5	259	92.1	66.7		12.4	0.01	619,6
26	Righia	6.7	8	3	12.8	0.4	24.4	19.3	1		2.5	0.02	71,4
27	Salsabil	7.95	25	5	27	4	125	10	21		11.4	<0.01	228,4
28	Sidi Rached	7.39	134.4	6.7	29.2	2.5	235	50	139		21.8	00	618,6
29	Soumam	7.21	114	32	71	2	293	78	196		19.2	<0.01	805,2
30	Tazliza	7.3	48	20	48	8	104	76	96		20	<0.01	420,0
31	Taya	7.3	47.3	12.5	55	1	-	110	82		17.4	0.01	325,2
32	Togi	7.46	73.4	19.3	36	1.8	-	43.8	28.9		5.9	<0.01	209,1

Khedidja, Mouzaïa, N'Gaous, SidiYacoub, Texanna, Toudja, Youkous) presented an ionic balance error greater than $|+/- 5\%|$ are excluded from the analysis base, including some for which the concentration of bicarbonates is absent. Thus, 15 bottled mineral waters out of the 24 collected were selected for study.

Eight bottled spring waters (Ain Bouglez, Ayris, Bir Salam, Ifren, Qniaa, Righia, Taya, Togi) showed an ionic balance error greater than $|+/- 5\%|$ are excluded from the analysis base, including some for which the concentration of bicarbonates is absent. Thus, 24 bottled spring waters out of the 32 collected were selected for study.

Piper Diagram

The chemical characteristics of the composition of the spring and mineral waters, on the basis of the

concentrations of major ions, were evaluated on the Piper diagram (Figure 1a and 1b).

We can observe on the diagram a variability of the chemical facies.

There are essentially two poles for mineral waters, a first pole of the chloride and sulfated calcium and magnesium type that can be seen for djemila, manbaa, thevest and sidiokba. The second pole is of the calcium and magnesium bicarbonate type that can be seen in the rest of the mineral waters studied.

Similarly, for spring water we can deduce that there are two types, water of the chlorinated and sulfated calcium and magnesium type that can be seen for ariaf, arwa, soumam, el kantara, ouwis, sidirached and tazliza. The second is of the calcium and magnesium bicarbonate type that can be seen in the rest of the spring waters studied.

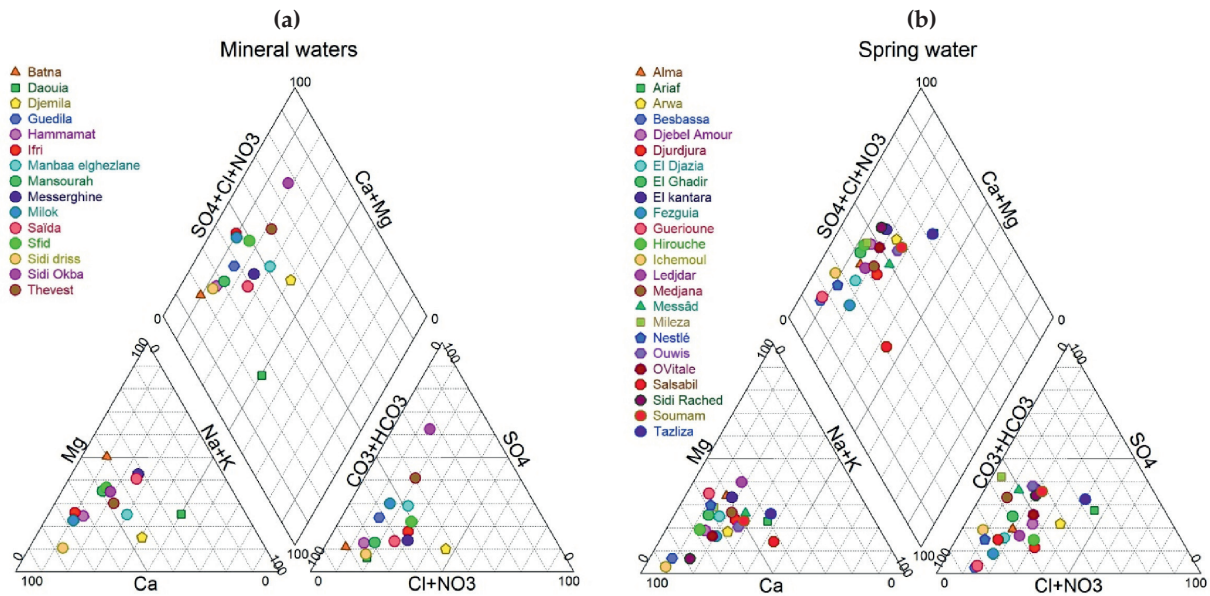


Fig. 1. Piper diagram of mineral waters (a) and springs (b) produced in Algeria

Overall, it can be seen that there is little diversity in the chemical facies of the waters; the vast majority of the waters studied are dominated by the calcium and magnesium bicarbonate type, which accounts for 72% of the waters studied. This is perfectly consistent with the results obtained by (Hazzab, 2011; Labadi and Hammache, 2016).

Statistical analyzes

Hierarchical Cluster Analysis (CAH)

The resulting dendrograms based on the similarity of the ten main ionic components grouped the fifteen brands of mineral waters and the twenty-four spring waters into similar groups.

Mineral water

Starting from the top of the dendrogram, we can count three groups with 64% similarity (Figure 2). The first group is made up of 9 brands: Batna, Daouia, Guedila, Ifri, Mansourah, Messerghine, Saïda, Sfid and Thevest. The second group includes three brands of bottled water: Hammamat, Milok and Sididriss. The third group includes three brands: Djemila, Manbaa and Sidi Okba.

The plot of the medians of the ionic components studied for the three groups (Figure 3), makes it possible to see that the first group includes the more water marks of water and which is characterized, by the average concentrations of the elements compared to the whole of the water marks studied except for the bicarbonates which reveal the highest

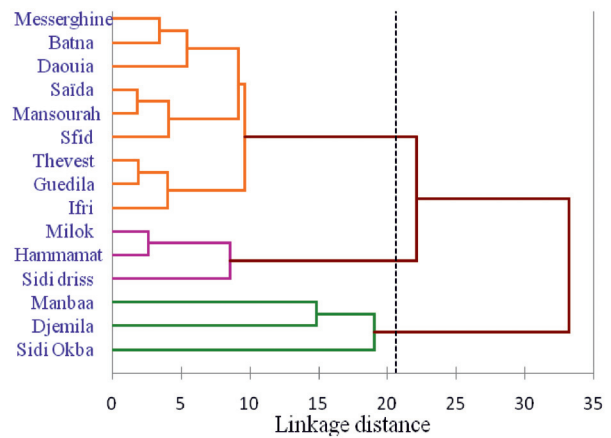


Fig. 2. Dendrograms for the hierarchical grouping of mineral water brands based on the main components of bottled waters.

concentration. The second group shows high concentrations of calcium, sodium, chlorides and sulfates. The third group is characterized by the lowest concentrations for all the elements studied. The medians of the parameters reveal a certain trend between the three groups.

Spring waters

As with mineral waters, the ascending hierarchical classification has made it possible to group spring waters into three classes with 54% similarity (Figure 4). The first group includes thirteen brands: Alma, Arwa, Djebel Amour, Djurdjura, El Djazia, El Ghadir, Fezguia, Guerioune, Hirouche, Ichemoul,

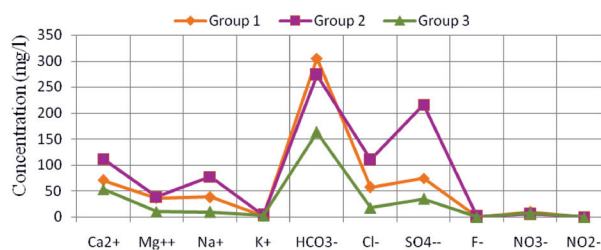


Fig. 3. Plot of the median values of the parameters for the three main groups of mineral waters determined using a hierarchical cluster analysis.

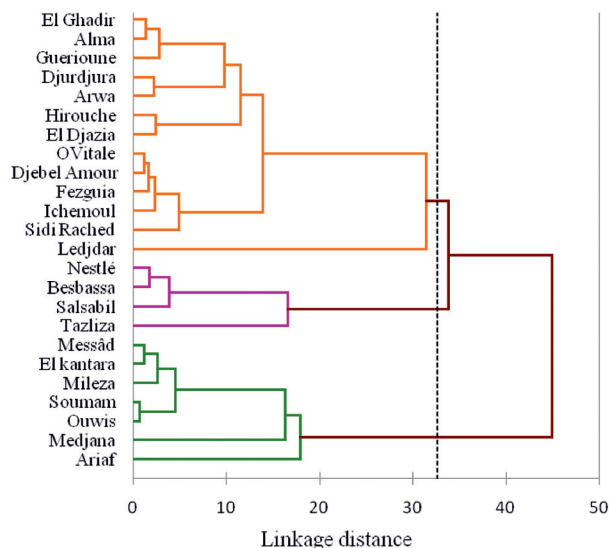


Fig. 4. Dendrograms for the hierarchical grouping of source water brands based on the main components of bottled waters.

Ledjdar, OVitale and SidiRached. The second, sevenbrands: Ariaf, El kantara, Medjana, Messâd, Mileza, Ouwis and Soumam. The third group contains four brands: Besbassa, Nestlé, Salsabil and Tazliza.

The plot of the medians of the ionic components studied for the three groups (Figure 5), shows that the first group includes the most brand water and

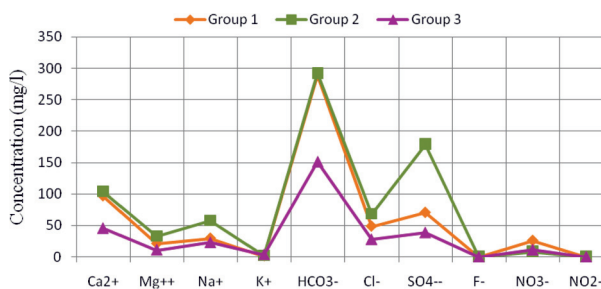


Fig. 5. Plot of the median values of the parameters for the three main groups of spring waters determined using a hierarchical cluster analysis.

which is characterized by the average concentrations of the elements compared to all of the brand waters studied with exception of bicarbonates and nitrates which show higher concentrations. The second group is characterized by the highest contents for the rest of the elements. The low concentrations of the ionic components are illustrated in the third group.

It should also be noted that the medians of the parameters reveal a certain trend between the three groups studied.

Comparison between mineral water and spring water

We can observe that some brands of water designated as natural mineral waters have a low mineralization for some brands such is the case for Sididriss, on the other hand some spring waters have high mineralization such as the waters of Medjana. In this regard, we conclude that the designation of bottled water as mineral water or spring water was done randomly, which is consistent with the study by (Bencheikh, 2021). This which is approved after the classification of all the samples into unknown groups depending on the similarity of their physico-chemical results using cluster dendrogram (Figure 6). This figure shows several similarities between mineral waters and springs, namely: (Thevest - Messâd), (Mansourah -

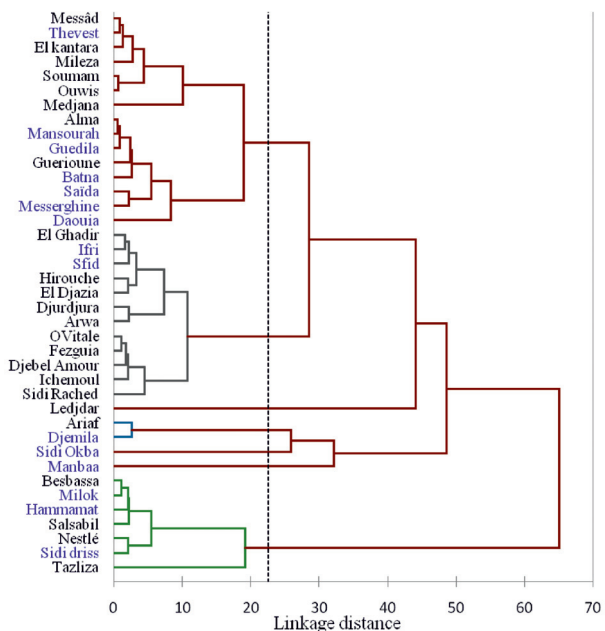


Fig. 6. Dendrograms for the hierarchical grouping of mineral and spring water brands based on the main components of bottled waters.

Alma), (Ifri - El Ghadir), (Djemila - Ariaef), (Milok - Besbassa) and (SidiDriss - Nestle).

Principal Component Analysis (PCA)

Figure 7 shows the layout of the 39 waters retained on the first two principal components. These waters have been scattered all over the graph showing some similarities between mineral and spring waters. It is also noted that, waters with low mineralization have been grouped on the left side of the graph on either side of the horizontal axis.

Variables with a correlation coefficient (r) value > 0.5 are considered significant. Important positive correlations are observed for some elements: Cl and Na ($r = 0.80$), SO_4 and Ca ($r = 0.67$), SO_4 and Mg ($r = 0.59$), HCO_3 and Mg ($r = 0.56$). Weak positive correlations were observed for: Cl and Ca ($r = 0.32$), HCO_3 and Ca ($r = 0.40$), SO_4 and Na ($r = 0.46$), Na and Mg ($r = 0, 40$), NO_3 and NO_2 ($r = 0.47$).

No significant correlation was obtained for K and F with the other variables, which corresponds well to their chemical composition which was low in all the brands of water studied.

PCA of water quality variables resulted in two principal components which account for about 46.05% of the total variance in the dataset and are characterized by a small positive contribution of Ca, Mg, Na, Cl and SO_4 (with a contribution of 40%, 42%, 45%, 39% and 44%, respectively). This component seems to be clearly dependent on the geological composition of the substrate, being located mainly in association with non-carbonate formations. The second principal component represents 15.66% of the total variance of the data and is characterized by a high negative contribution in NO_3 and NO_2 (with a contribution of 63%, and 68%, respectively).

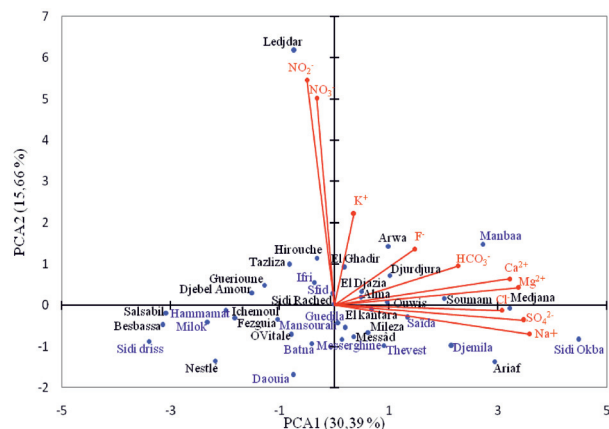


Fig. 7. Graph of observations and correlations between major elements

CONCLUSION

Many people prefer to drink bottled water than tap water, for a number of reasons. For some, because of the bad taste, smell or color of tap water. Others worry about their health and view bottled water as a natural, purer and healthier alternative.

Local bottled water production has developed considerably over the past three decades in Algeria. Thus and faced with the great diversity of brands of water marketed (origin, chemical composition, price, etc.), the consumer is sometimes confused as to the choice of the one that suits him.

This work has set itself the objective of distributing the waters most used by consumers, using a chemical classification and statistical tools, into groups or categories of waters that are more or less homogeneous from the point of view chemical element content.

The ion balance check on all the water collected showed that 30% of the water presented an acceptable charge balance error. The retained water was subjected to chemical and statistical distribution methods.

The chemical classification according to the Piper diagram led to the identification of two main chemical types: 28 % of the waters studied are of the chlorinated and sulphated calcium and magnesium type and 72 % of the calcium and magnesium bicarbonate type.

The hierarchical cluster analysis made it possible to group the so-called spring waters into three groups (Alma, Arwa, Djebel Amour, Djurdjura, El Djazia, El Ghadir, Fezguia, Guerioune, Hirouche, Ichemoul, Ledjdar, OVitale and SidiRached - Ariaef, El kantara, Medjana, Messâd, Mileza, Ouwis and Soumam - Besbassa, Nestlé, Salsabil and Tazliza) and so-called mineral waters in three groups (Batna, Daouia, Guedila, Ifri, Mansourah, Messerghine, Saïda, Sfid and Thevest - Hammamat, Milok and Sididriss - Djemila, Manbaa and Sidi Okba). The analysis of all the brands studied together showed that the designation of bottled water as mineral water or spring water was done randomly.

The representation of mineral waters by the PCA shows convergences towards the results obtained by the CAH, namely the resemblance of some water brands, but it does not seem particularly useful since the first two components represent only 46% of the variance total.

Finally, we can say that the distribution resulting from this study is based on the quantitative

composition of the major chemical elements of the waters studied, and does not give the privilege to any brand of water. The main exploitable result at this level is to allow consumers to expand the range of choices of brands of waters with similarities in terms of chemical composition.

CONFLICTS OF INTEREST

The authors declare no conflicts of interest regarding the publication of this paper.

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