Geochemical Impacts of the Physical Parameters and Heavy Metals Risk of the Drinking Bottled Water Produced and Marketed in Baghdad City, Iraq

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Abstract
Physico-chemical parameters of fourteen bottled water brand produced and marketed in Baghdad Capital City with two tap water samples were environmentally assessed. The results are compared with corresponding guidelines of Iraqi Standards and the World Health Organization.The mean values of physical and aggregate properties of the tested waters are; pH, 6.5 and 6.9, EC,192 and 555 µS/cm at 25◦C, TDS, 92 and 275 mg/L, Total hardness as CaCO3, 36.97 and 143 mg/L and alkalinity as HCO3-1, 44.85 and 66 mg/L. The mean concentration of major ionic composition of the tested waters are; Cl^{-1}, 34.8 and 40.55 mg/L, $SO4^{-2}$ 35.58 and 126.5 mg/L, Ca^{+2} 7.67 and 10 mg/L, Mg^{+2} 4.3 and 10 mg/L, K^{+1} 0.639 and 1.35 mg/L and Na^{+1} 9.4 10 mg/L. These properties show increasing in the tape water than those of bottled waters.Classification based on TDS suggests very low to little low mineral concentrations and total hardness suggests soft to moderately hard waters. Piper diagram reveal bicarbonate-sulfate water type suggests originating from carbonate/gypsiferous sediments.Geochemical concentrations of heavy metals analysis in bottled waters are less than those in tap waters except B and Pb concentrations. Arsenic concentration shows value lies in between polluted and unpolluted level, which probably comes from damages of tap and waste waters networks. Three water samples show high concentration of sulfate ions, suggest originated in gypsiferous sediments.The physicochemical parameters of bottled and tap waters are coincide with guidelines of World Health Organization and Iraqi Standards, which indicates potability without risks.
Keywords: Bottled water, tap water, physico-chemical parameters, Piper diagrams, heavy metals, ICP-MS.

1. Introduction

Consumption of the Bottled waters is increasing steadily worldwide during the last three decades from 130,956 million liters in 2002 to 188,777 million liters in 2007. Thus, the average annual consumption rate is 28.8 L/capita, European Union consumption is 104.2 L/capita, and in USA are 110.9 L/capita (European Federation of Bottled Water 2006; Beverage Marketing Corporation 2008). In recent years, the production of bottled water was increases for over 5,000 brands in the worldwide (Güler 2007).

Drinking waters are packed in bottles or containers adding optionally safe and suitable antimicrobial agents. Fluoride may add within established quality standards (FDA 2008). However, despite its abundance, good quality drinking water is not readily available to human drinks (Onweluzo and Akuagbazie 2010). Safe and good quality drinking water is essential for human health and physiology for continuous lives (Petraccia et al. 2006; Karavoltsos et al. 2008; Alemdar et al. 2009; Krachler and Shotyk 2009). The human health needs 3 liters/average for weight of 53 kg - 63 kg, in the liquids and food daily (Wardlow et al. 2004).

Literature revealed that some constituents in bottled water may be in violation of action levels for various parameters (Karamanis et al. 2007). WHO /Food and Agriculture Organization regulates the bottled and mineral waters (CAC 2001). Other countries sited acceptable standards for bottled water (like Iraq), typically in harmonization with broader regulatory agencies such as WHO, USEPA and the Council of European Economic Community Directive.

The quality of drinking waters was deteriorated during recent years due to high levels of pollutions, which caused serious health hazard (Warburton 1993; Nkono and Asubiojo 1997; Ikem et al. 2002; Turk and Alp 2010). Concentrations of toxic chemicals and metals e.g. cadmium, mercury and lead, in drinking waters may cause defect particularly cardiovascular, kidney-related disorders, neurocognitive effects and cancer diseases (Al -Saleh and Al- Doush 1998; IKem et al. 2002).

The indiscriminate discharge of industrial effluents leached to the ground water aquifers, lead to chemical pollution of natural water bodies (Ikem et al. 2002). The rapid increases in the production of bottled water and prompts of market concern about the quality of bottled water produced in Iraq. Waste materials of wars and damages of tap and waste water networks in Baghdad City comprises big sources of pollution for the soil and underground waters through the leaching of rainfalls.

Therefore, quality assessment of the physico-chemical parameters impact was carried out for validity of the drinking bottled waters produced in Baghdad City for human consumption. This includes the analysis of the most important toxic heavy metals that make human health risk. Moreover, two samples of tap waters were collected from each side of Baghdad City to assess validity and compare with bottled water. Along with the results of these parameters and heavy metals content are compared with prescribed Iraqi and World Health Organization standards guideline.

2. Materials and methods

Fourteen (14) brands of drinking bottled waters were collected during summer 2013 from randomly selected groceries and supermarkets from Baghdad City-Iraq. These bottled waters are produced from drilled-wells from underground water aquifers, which are lying within thick sediment layers of Tigres River within the Mesopotamian Plane. The level depths of the underground water aquifers are varied from location to another, most of them are of shallow depths. The drinking water brands were purchased in duplicate polyethylene terephthalate (PET) containers of sizes 0.5, 1.0, and 20 liter. All these samples were stored at ambient conditions (23–25◦C) prior to the concerned analyses. General information about the chemical composition (e.g. pH, TDS, Na^+, K^+, Mg^{+2}, Ca^{+2}, Cl^-, $SO4^{-2}$ and $HCO3^-$ parameters with expiry dates) are listed on labels on the bottles. To keep the brand names anonymous, water samples given numerical codes and this convention was used throughout the study.

The physical parameters of the water samples were measured using the portable meter device include pH, EC, TDS and TH parameters. The major nonmetallic inorganic constituents include; (Na^+, K^+, Mg^{+2}, Ca^{+2}, Cl^-, $SO4^{-2}$ and $HCO3^-$) and heavy metals include; As, B, Ba, Cd, Cr, Cu, Co, Fe, Hg, Mn, Mo, Ni, Pb, Se, U, V and Zn were determined by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) in the ALS Laboratory in Spain.

Two tap water samples were collected from the two sides of Baghdad City (Al-Rusafah and Al-Karkh) representing the wide usage of drinking waters in the human population. It is important to mention that all the analytical tests followed the Standard Methods (APHA et al. 2005) and/or the USEPA accepted reference methods (Nelson 2003). All analyzed data is compared with WHO (2008) and ISQ (2009) standards to assess the expected environmental pollution.

3. Results and discussion

3.1 Quality assessment of the physico-chemical parameters

The results of the analyzed physico-chemical parameters for drinking bottled waters are summarized in Table (1&2), together with corresponding guidelines of World Health Organization (2008) and Iraqi Standards (2009) for comparison. The physical and aggregate properties of the tested bottled waters are listed in the following;

pH is range between 6.1 and 7.1 (mean = 6.5), EC is 10 and 310 µS/cm at 25◦C (mean 192 µS/cm at 25◦C), TDS is 0.0 and 150.0 mg/L (mean =92 mg/L), total hardness (TH) is 0.55-65 mg/L as CaCO3 (mean = 36.97 mg/L as CaCO3) and the total alkalinity (as CaCO3) is 15-81 mg/L as $HCO3^{-1}$ (mean = 44.85 mg/L). The major anions and cations content of the tested bottled waters are; Cl^{-1} is 1 and 187 mg/L (mean = 34.8 mg/L), $SO4^{-2}$ is 5 and 79.4 mg/L (mean = 35.58 mg/L), $Ca+^2$ is 0.2 and 10 (mean = 7.67 mg/L), $Mg+^2$ is 0.012 and 9.79 mg/L (mean = 4.3 mg/L), 0.06, K^{+1} is 1.45 mg/L (mean = 0.639 mg/L) and Na^{+1} is 4.3 and 10 mg/L (mean = 9.4 mg/L) for.

The physical and aggregate properties of the tested tap waters range are; pH is 6.8 and 7.0 (mean= 6.9), EC is 500 and 610 µS/cm at 25◦C (mean 555 µS/cm at 25◦C), TDS is 250 and 300 mg/L (mean =275 mg/L) for, total hardness as CaCO3 is 143-143 mg/L (mean = 143 mg/L) and total alkalinity is 66-66 mg/L as $HCO3^{-1}$ (mean = 66 mg/L). The major ionic composition of the tested tap waters are; Cl^{-1} is between 32.8 and 48.3 mg/L (mean = 40.55 mg/L), $SO4^{-2}$ is112 and 141 mg/L (mean = 126.5 mg/L), Ca^{+2} is 10 and 10 (mean = 10 mg/L), $Mg+^2$ is 10 and 10 mg/L (mean = 10 mg/L), $K+^1$ is 1.28 and 1.43 mg/L (mean = 1.35 mg/L) and $Na+^1$ is 10 mg/L (mean = 10 mg/L).

Samples no. 4B, 7B and 14B shows abnormal concentrations of sulfate ions (SO^{-2}), these are; 76.1, 79.4 and 54.2 respectively (Table 2). While the labeled data upon the bottled are differ (Table 3). This suggests that the source of these waters is produced from aquifer situated in gypsiferous or gypcrete sediments. It is important to mention that the soil in the west and southern parts of Baghdad is composed of gypcrete soil (Geder 2004; Hassan 2014).

EC reflects the total mineralization of the tested bottled waters. According to WHO (2008) the EC in the natural mineral waters are categorized into low mineral content e.g. oligomineral waters (Feru 2004). The analyzed water quality shows acceptable limits to respective guidelines of the World Health Organization (WHO 2008) and the Iraqi Standards (IQS 2009).

Because of unknown origin of the bottled waters, which is generally produced from drilled water wells in Baghdad City and surroundings, and because of great damages of the tap and waste waters networks e.g. the big sources of pollutions, concentrations of major ions of the analyzed waters were evaluated using Piper diagram (Fig 1) generated by the GW Chart software v. 1.19.0.0 (Winston 2000). It is readily seen that the investigated bottled waters fall in the field of Earth alkali water with prevailing sulfate probably originating from sediments rich in gypsiferous and carbonates with little salt patches.

3.2 Classification of tested bottled and tap water

European Union (EU) mineral water directive (Vander Aa 2003) was used to classify the water according to TDS and total hardness parameters (Table 4). The classification systems used to identify the chemical similarities and/or differences between the bottled water brands. For the case of brands 1B and 10B fall in the class “very low mineral concentration” (TDS<50 mg/L), while the other brands fall in class “low mineral concentration” (TDS between 50-500 mg/L) (Mihayo and Mkoma 2012).

3.3 EC, TDS, pH

The chemical content of the bottled waters is a function of the origin and migration of waters, consequently the rock composition or sediments of the aquifer. The chemical content is controlled by certain parameters such as CO2 concentration, redox conditions and the type of adsorption complexes (Birke et al. 2010). The levels of EC and TDS in bottled water brands are shown in Table (2) compared with the Iraqi Standards and WHO limits. The ranges for these parameters are 10-610 µS/cm for EC with mean of 187.2 µS/cm and 0-300 mg/L for TDS with mean of 95.2 mg/L. There are variations between EC and TDS values of the bottled water samples, which depend on the origin of the waters, residence time, atmospheric conditions and the treatment or purification process (Mihayo and Mkoma 2012). The pH of all brands was around neutral, which is ranging from 6.1-7.1 with mean value 6.5.

4. Heavy metals concentrations

The majority of diseases and health risk are caused morbidity and mortality from water related pollution by heavy metals (Shayo et al. 2007). The mean concentrations of the heavy metals in the analyzed bottled and tap waters are shown in Table (5). All concentrations of heavy metals are lying within the guideline standard limits of the World Health Organization (WHO 2008) and Iraqi Standard (IQS 2009).

The heavy metals in bottled water brands are ranked as B> Fe >Ba> As, Se > Zn> Cr> V> Mo> Cu> Ni> Pb> Hg> Mn> Cd, Co> U, while in tap water samples ranked as Fe >B >Ba> Zn> As, Se> Cr> V> Mo >Cu >Ni> U> Mn> Hg> Pb> Cd, Co.

The mean concentrations of B and Pb in bottled water are higher than tap water, while the others As, Ba, Cd, Cr, Cu, Co, Fe, Hg, Mn, Mo, Ni, Se, U, V and Zn are within or lower than tap water Table (5). The results of this study shows that mean lead (Pb) level was slightly higher (0.27 µg/L) than tap water (0.2 µg/L), but within the levels of the guidelines. This is disagree with El-Harouny et al. (2008) who found that the lead levels in tap water from Dakhlia governorate-Egypt is higher than total bottled water. The most probable source of Pb pollution is coming from the crude oil combustion used as a fuel in the electrical power plants, which are distributed around Baghdad City. The irrigation and rainfall leaching waters are responsible for Pb concentrations in underground water aquifers and consequently to the produced bottle waters. Another source of Pb and B ions is the wide uses of fertilizers in the scattered farms and agriculture areas around Baghdad City. The mean Boron level (189 µg/L) is higher than the tap water (65 µg/L), but within the levels of the guidelines. No previous study on boron level in bottled water was recorded.

Arsenic concentrations are ≤10 µg/L in all brands of bottled and tap water samples, which lies in the critical limited between polluted and polluted level according to the WHO (2008) standards. Arsine is the most toxic form as arsenates and organic arsenic compounds (WHO 2008). The most probable source of Arsenic (As) in waters is the waste waters, which comes from mixing of tap and waste waters in underground Baghdad City. The great damages of both tap and waste waters networks due to a series of wars since 1991. Moreover, both networks are expired out of time.

All other mean levels of the heavy metals in the drinking bottled and tap waters are below the levels of the World Health Organization (WHO 2008) and IQS (2009) water quality standards. The levels of Ba, Cd, Cr, Cu, Co, Fe, Hg, Mn, Mo, Ni, Se, U, V and Zn are within or lower than the mean level of the tap water. This agreed with most studies e.g. El-Harouny et al. (2008) found that the mean concentrations of the heavy metals (Pb; Cd; As; g Na; Ca; Mg; Zn and Se) in drinking water of Dakhlia governorate-Egypt are in the acceptable levels of the World Health Organization (WHO 2004). While the tap water shows slightly higher levels of lead, arsenic, cadmium, zinc, and selenium. David et al. (2013) found that the concentrations of Mg, Ca, Mn, Fe, and Cd in the samples were low when compared with World Health Organization (WHO 2005) standards. Yabanli (2012) found that the levels of Al, Cr, Cu, As, Cd, and Pb in bottled water sold in Izmir-Turkey are within the concentration levels of WHO and international standards.

5. Conclusion

Fourteen bottled water brands and two tap water samples were analyzed to assess the quality of physico-chemical parameters. Results show various characteristics aggregates, yet the majority met the various national and international bottled water standards. However, the quality of physico-chemical of the studied water samples was variable, probably depends various factors e.g. natural environment, aquifer composition and the type of treatment/purification technique(s) in the production. The chemical quality of the studied waters depends essentially on the source and chemical composition of the origin of the waters.

For the unknown origin of the waters type generated Piper diagrams shows that the majority of the tested waters are of magnesium carbonate with little sulfate. These suggest that the aquifer origin situated in carbonate with some gypsum and/or gypsiferous sediments. Comparison of the results with reported label values for the bottled waters indicate good agreement with stated pH values as well as Na^{+1}, K^{+1}, Ca^{+2}, Mg^{+2}, $HCO3^{-1}$, Cl^{-}, and $SO4^{-2}$. Three bottled water samples show abnormal concentration of sulfate ions suggest water is originated in aquifer lies in gypsiferous and/or gypcrete sediments.

Investigation of chemical quality of bottled water samples produced in Baghdad City shows that all brands of bottled water were potable and fit for human consumption and gave a good indication of the present state of metal contamination of tested water, which is at very low levels.

In general, all elemental concentrations are within the acceptable limits of the World Health Organization (2008) and f Iraqi Standards (2009) for drinking water indicating the potability and save bottled waters.

Appropriate monitoring programs should be established to ensure that the chemical quality of drinking water remains within acceptable national standards and to maintain water safety for public use. Reduction of metals which have cumulative toxic hazards in drinking water is a necessity.

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Table 1. Physical and aggregate properties of the investigated bottled and tap waters from Baghdad City.

Brand code	**pH (at 25° C)**	**TDS (mg/L) HCO3-1** µS/cm	**EC (µS/cm at 25° C)**	**Bicarbonate alkalinity (mg/L CaCO3)**	**Total hardness (mg/L CaCO3)**
1B	7	0	10	15	0.55
2B	6.5	90	180	31	43.5
3B	6.5	50	120	35	38.7
4B	6.5	150	310	81	59.9
5B	6.1	140	290	18	10.9
6B	6.5	150	310	15	11.6
7B	6.1	80	180	79	65.23
8B	6.5	150	310	48	45.4
9B	6.6	70	140	42	41.23
10B	6.7	30	70	37	9.3
11B	6.5	90	180	46	45.13
12B	6.5	120	240	77	53.4
13B	6.5	50	110	42	38.4
14B	7.1	120	240	62	54.4
Range	**6.1-7.1**	**0-150**	**10-310**	**15-81**	**0.55-65.23**
Mean	**6.55**	**92.142**	**192.142**	**44.857**	**36.97**
1TK	6.8	250	500	143	66
2TR	7	300	610	143	66
IQS,2009	**6.5-8.5**	**--**	**1000**	**---**	**500**
WHO, 2008	**6.5-9.5**	**2500**	**600**	**250**	

Table 2. Concentrations (mg/L) of the major ions of investigated waters from Baghdad City.

Brand code	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	Cl^{-}	SO_4^{2-}	HCO_3^{-}
1B	0.2	0.012	4.300	0.06	1	5	15
2B	10	4.470	10	0.510	24.5	43.6	31
3B	10	3.340	9.300	0.360	10.4	26.5	35
4B	10	8.590	10	0.680	27.2	76.1	81
5B	3.1	0.837	10	1.180	104	11.3	18
6B	1.7	1.820	10	0.660	187	21	15
7B	10	9.790	10	0.8	28.2	79.4	79
8B	10	4.960	10	1.450	16.2	40.5	48
9B	10	3.950	8.400	0.380	10.2	34.6	42
10B	2.4	0.809	10	0.490	14.4	12.5	37
11B	10	4.920	10	0.680	17.8	34	46
12B	10	6.930	10	0.7	17.2	42.2	77
13B	10	3.250	10	0.410	9.83	17.3	42
14B	10	6.910	10	0.590	19.5	54.2	62
Range	**0.2-10**	**0.012-9.79**	**4.3-10**	**0.06-1.45**	**1-187**	**5-79.4**	**15-81**
Mean	**7.67**	**4.327**	**9.428**	**0.639**	**4.816**	**35.586**	**44.857**
1TK	10	10	10	1.280	32.8	112	143
2TR	10	10	10	1.430	48.3	141	143
IQS,2009	**150**	**100**	**200**	**--**	**350**	**--**	**--**
WHO, 2008	**--**	**--**	**200**	**---**	**250**	**250**	

Table 3. Labeled physical and chemical parameters on the large bottled water of 20 L capacity.

Water type	**Chemical parameters**										**Physical parameters**		
	Cations				**Anions**								
	Ca^{+2}	**Mg^{+2}**	**Na^{+1}**	**K^{+1}**	**$HCO3^{-1}$**	**$NO3^{-1}$**	**$SO4^{-2}$**	**F^{-1}**	**Cl^{-1}**	**B^{-1}**	**pH**	**TDS**	**TH**
Al-Barq	13.0	2.0	11.0	0.6	85	1.0	18.2	1.0	3.1	-	7.3	80	43
Al-Rabe'a	14.4	3.0	12.3	1.5	-	2.5	28	0.9	17.5		7.2	60	40
Jana	50	20	40	-		-	50	-	0.0	-	7.4	150	-

Table 4. The classification of the tested drinking bottled and tap waters based on EU mineral water directive (Mihayo and Mkoma, 2012).

Brand	**TDS**	**EU class**	**TH**	**Water class**
1B	0	Very low mineral concentration	0.55	soft water
2B	90	Low mineral concentration	43.5	soft water
3B	50	Low mineral concentration	38.7	soft water
4B	150	Low mineral concentration	59.9	moderately hard water
5B	140	Low mineral concentration	10.9	soft water
6B	150	Low mineral concentration	11.6	soft water
7B	80	Low mineral concentration	65.23	moderately hard water
8B	150	Low mineral concentration	45.4	soft water
9B	70	Low mineral concentration	41.23	soft water
10B	30	Very low mineral concentration	9.3	soft water
11B	90	Low mineral concentration	45.13	soft water
12B	120	Low mineral concentration	53.4	moderately hard water
13B	50	Low mineral concentration	38.4	soft water
14B	120	Low mineral concentration	53.4	moderately hard water
1TK	250	Low mineral concentration	66	moderately hard water
2TR	300	Low mineral concentration	66	moderately hard water

Table 5. Concentration of the heavy metals in investigated bottled and tap water in (µg/l).

Brand Code	As	B	Ba	Cd	Cr	Cu	Co	Fe	Hg	Mn	Mo	Ni	Pb	Se	U	V	Zn
1B	< 10	90	0.11	0.1	1	0.3	0.1	20	0.2	0.1	1	0.5	0.2	10	0.01	1	2
2B	< 10	80	6.28	0.1	1	0.3	0.1	20	0.2	0.1	1	0.5	0.2	10	0.05	1	4
3B	< 10	40	8.06	0.1	2	0.3	0.1	20	0.2	0.1	1	0.5	0.2	10	0.04	1	2
4B	< 10	50	23.5	0.1	4	0.8	0.1	40	0.2	0.2	1	0.5	0.2	10	0.15	3	5
5B	< 10	900	0.94	0.1	1	1.4	0.1	20	0.2	0.2	1	0.9	0.2	10	0.01	1	7
6B	< 10	940	0.21	0.1	1	0.3	0.1	20	0.2	0.1	1	0.5	0.2	10	0.01	1	4
7B	< 10	60	21.9	0.1	4	0.7	0.1	70	0.2	0.1	1	0.5	0.2	10	0.16	3	3
8B	< 10	60	11.55	0.1	2	0.3	0.1	40	0.2	0.1	1	0.5	0.2	10	0.06	2	2
9B	< 10	50	10.8	0.1	2	0.4	0.1	20	0.2	0.3	1	0.5	0.2	10	0.07	2	3
10B	< 10	150	1.37	0.1	1	0.3	0.1	20	0.2	0.1	1	0.5	0.2	10	0.03	1	5
11B	< 10	80	8.63	0.1	2	0.3	0.1	20	0.2	1.1	1	0.5	0.2	10	0.08	2	51
12B	< 10	50	17.8	0.1	4	0.3	0.1	30	0.2	0.1	1	0.5	0.2	10	0.12	3	4
13B	< 10	50	7.93	0.1	2	0.3	0.1	20	0.2	0.1	1	0.5	0.2	10	0.06	2	2
14B	< 10	50	17.85	0.1	4	5.4	0.1	30	0.2	0.1	1	0.5	1.3	10	0.12	2	5
Range	< 10	40-940	0.11-23.5	0.1	1-4	0.3-1.4	0.1	2-70	0.2	0.1-0.3	1	0.-0.9	0.2	10	0.0-0.16	1-3	2-51
Mean	< 10	131.81	15.58	0.1	3.54	0.62	0.1	36.8	0.2	0.17	1	0.57	0.25	10	0.09	2.45	7
1TK	< 10	20	21.5	0.1	5	0.3	0.1	70	0.2	0.1	1	0.6	0.2	10	0.13	3	2
1TR	< 10	140	10.45	0.1	2	0.3	0.1	50	0.2	0.1	1	0.5	0.2	10	0.1	1	2
IQS, 2009	10	500	700	3	50	100		300	1	100	--	20	10	10	--		3000
WHO2008	10	500	700	3	50	200		300	6	400	70	70	10	10	15		3000

**B: Baghdad city, T: tap water*

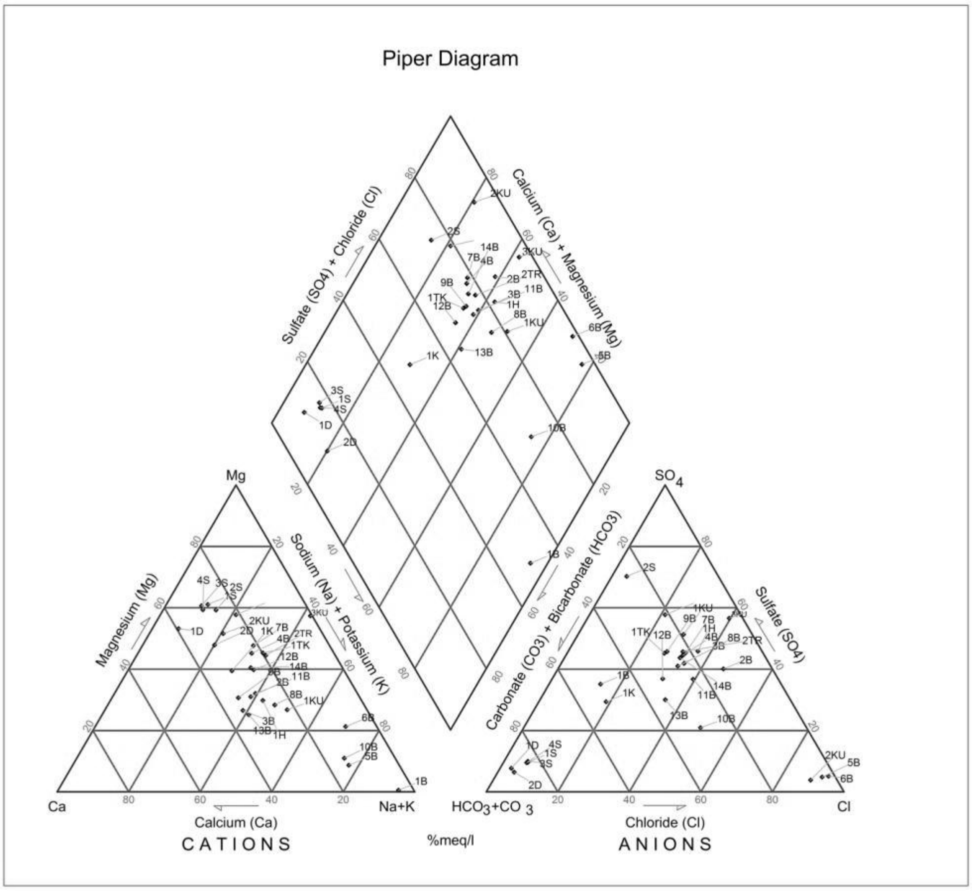

Fig 1. Application of Piper diagrams for investigated bottled waters.