

Trace and Major Elements in Iranian Bottled Mineral Water: Effect of Geology and Compliance with National and International Standards

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1. Introduction

Mineral bottled water, as a safe source of drinking water, has gained a universal popularity in the last few decades (Cidu et al., 2011). Mineral water is the most common type of bottled water (Doria, 2006), which contains essential element cations and anions derived from lithological sources (Biddau et al., 2019). As many studies on bottled mineral water have shown, trace elements, above certain levels, can be detrimental to human health (Güler et al., 2002; Birke et al., 2010a; Bityukova et al., 2010; Cicchella, et al. 2010; Frengstad et al., 2010; Petrović, et al., 2010; Reimann and Birke, 2010; Smedley, 2010; Banks et al., 2015). Due to the fact that mineral water has its origin from groundwater resources, the influence of regional and in particular aquifer geology on their chemical composition is expected (Brenčič, et al. 2010; Platikanov et al., 2013). Chemical composition of groundwater reflects the local geology of spring source area and the geochemistry of the rocks in contact with the water (Daniele et al., 2019). Groundwater in aquifers may interact with various minerals and become enriched or depleted in some elements (Peh et al., 2010). In Iran, the demand for bottled mineral water consumption has been growing extremely fast in the last few years. However, the detailed chemistry of this vulnerable resource has hitherto received little attention (Samadi et al., 2009; Miranzadeh et al., 2015; Kermanshahi et al., 2010; Salehi et al., 2014). To determine the quality of Iranian bottled mineral water in relation to the contribution of various lithological units, we studied 36 best-selling brands of bottled mineral water throughout the country, and compared the results with the threshold limit values.

2. Methods and Materials

2.1. Characteristics of water sources

The bottled mineral water samples were selected from eighteen provinces located in four different lithological units of Iran, namely Zagros, Alborz, Central Iran and Kopet Dagh (Fig. 1). Groundwater from the

spring sources in Chahar Mahal Bakhtiari, Ilam, Kermanshah, Fars and Kohgiluyeh provinces is thought to be influenced by Zagros sedimentary rocks. This unit is dominantly composed of carbonate rocks with interlayers of shale, sandstone, chert, anhydrite and gypsum. The brands of bottled mineral water, sold in the markets of Tehran, Alborz, Ardebil, Zanjan, Golestan, Guilan and Mazandaran provinces, pass through several structural subdivisions of Alborz with different lithological components of sedimentary rocks as well as andesitic to basaltic volcanic rocks (Stöcklin, 1968). Even though Central Iran is dominated by metamorphic and magmatic host rocks, the selected Kerman water has its source in Oligo-Miocene carbonates of this unit. The source of the bottled mineral water produced in Khorasan Razavi province is located in a thick sequence of limestone and dolomite of Kopet Dagh basin in north-eastern Iran.

The number of studied samples per geological unit is proportional to the number of producers in each

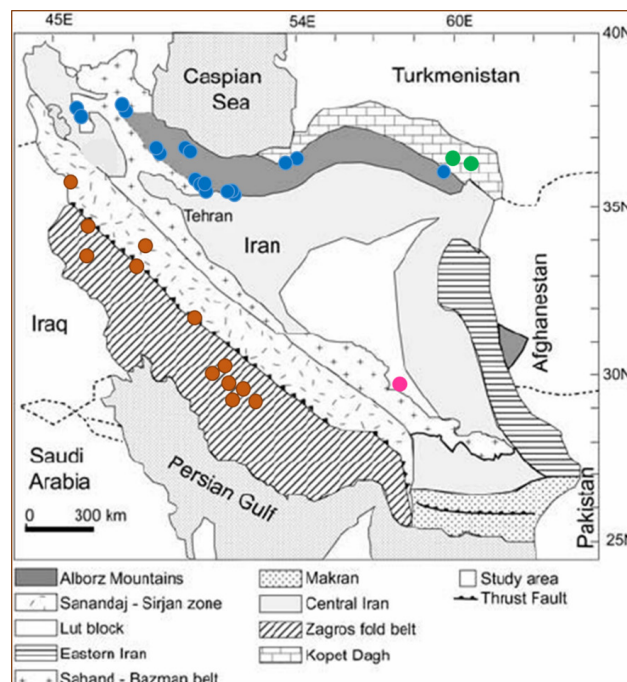


Figure 1: The location of water samples on the map of structural zones of Iran (Stöcklin 1968). Red circle = samples from the Zagros orogeny (folded Zagros and Sanandaj-Sirjan), blue circle = samples from Alborz Mountain and its extension, green circle = samples from Kopet-Dagh, and pink circle = samples from Central Iran unit.

	Unit	Techn.	D.L.	Mean	Median	Std. Dev	Min	Max	25ile	50ile	75ile	Spread	ISIRI	WHO
pH	-	-	0,01	7,81	7,86	0,2	7,27	8,12	7,72	7,86	7,93	1,12	6.5–9	
EC	µS/cm	-	1	366	347	137	95	742	265	347	419	8		
TDS	mg/l	-		238	226	89	62	482	172	226	272	7,8	1500	
TH	-	-		167	162	71	33	325	125	161	217	9,8	500	
Macro Elements														
Ca	mg/l	ICP-OES	0,1	47,1	46,2	20,8	0,1	95,2	36,4	46,2	59,1	952	300	
K	mg/l	ICP-OES	0,1	0,7	0,7	0,5	0,1	3	0,4	0,7	0,8	30		
Mg	mg/l	ICP-OES	0,01	12,05	9,89	7,97	0,04	36,5	6,66	9,89	17,85	912		
Na	mg/l	ICP-OES	0,1	13,3	8,6	13,8	0,7	59,7	4,4	8,6	19,5	85	200	50
Mn	µg/L	ICP-MS	0,01	0,1	0,05	0,12	0,01	0,54	0,03	0,05	0,14	54	400	
SiO ₂	mg/l	ICP-OES	0,1	10,7	9,1	6,9	0,1	33,7	6,2	9,1	14,1	337		
Sr	mg/l	ICP-OES	0,001	0,427	0,339	0,361	<0,001	1,85	0,234	0,339	0,502	3700		
Br	mg/l	IC	0,003	0,023	0,017	0,042	0,003	0,251	0,005	0,017	0,028	83		
Cl	mg/l	IC	0,01	13,31	7,6	14,76	0,86	59,4	2,52	7,6	23,72	69	400	5
F	mg/l	IC	0,01	0,157	0,128	0,122	0,003	0,696	0,091	0,128	0,217	232	4	1,5
HCO ₃	mg/l	Titration	0,1	172	172	74	3	322	140	172	214	107		
NH ₄	mg/l	Photometry	0,01	<0,01	<0,01	-	<0,01	<0,01	<0,01	<0,01	<0,01	-		
NO ₂	mg/l	IC	0,003	<0,003	<0,003	<0,003	<0,003	0,01	<0,003	<0,003	<0,003	-	20	3
NO ₃	mg/l	IC	0,001	9,34	5,78	8,68	0,16	36,2	3,7	5,78	12,68	226	50	50
PO ₄	mg/l	ICP-OES	0,03	0,02	<0,01	0,16	<0,01	0,87	<0,01	<0,01	<0,01	-		
SO ₄	mg/l	IC	0,01	24,66	14,6	31,9	0,66	130	6,93	14,6	29,35	197	400	
Micro and Trace Elements														
Al	µg/l	ICP-MS	0,01	1,5	0,9	1,5	0,1	5,5	0,5	0,9	1,7	55		200
As	µg/L	ICP-MS	0,01	1,98	0,32	9,59	0,01	57,92	0,21	0,32	0,44	5792	10	10
B	µg/L	ICP-MS	0,01	64,1	20,1	138,2	4,8	692	11,2	20,1	52,9	144		2400
Ba	µg/L	ICP-MS	0,01	29,5	16,28	40,71	0,02	237,34	8,68	16,28	39,98	11867	700	700
Cd	µg/L	ICP-MS	0,001	<0,001	<0,001	0,01	<0,001	0,03	<0,001	<0,001	<0,001	60	3	3
Co	µg/L	ICP-MS	0,001	0,017	0,014	0,009	0,006	0,05	0,011	0,014	0,02	8,3		
Cr	µg/L	ICP-MS	0,01	0,96	0,59	1,24	0,02	6,7	0,35	0,59	1,05	335	50	50
Cu	µg/L	ICP-MS	0,01	5,54	0,23	0,79	0,01	3,01	0,08	0,23	0,65	301	2000	2000
Fe	µg/l	ICP-MS	0,1	0,3	0,2	0,3	<0,1	1,6	0,1	0,2	0,3	32		
Hg	µg/L	ICP-MS	0,01	<0,01	<0,01	0,01	<0,01	0,02	<0,01	<0,01	0,01	4	1	6
I	µg/L	ICP-MS	0,1	6,3	4,5	5,6	0,2	22	2,9	4,5	6,4	110		
Li	µg/L	ICP-MS	0,1	5,4	2,7	9,5	0,1	56,7	1	2,7	6,1	567		
Mo	µg/L	ICP-MS	0,01	1,01	0,75	0,95	0,01	3,83	0,29	0,75	1,37	383		
Ni	µg/L	ICP-MS	0,01	0,26	0,16	0,3	0,07	1,71	0,13	0,16	0,21	24	70	70
Pb	µg/L	ICP-MS	0,01	0,04	0,01	0,08	<0,01	0,4	<0,01	0,01	0,03	80	10	10
Rb	µg/L	ICP-MS	0,01	0,42	0,26	0,8	0,02	4,71	0,1	0,26	0,39	235		
Sb	µg/L	ICP-MS	0,001	0,329	0,243	0,201	0,141	0,988	0,186	0,243	0,463	7	5	20
Se	µg/L	ICP-MS	0,01	0,49	0,29	0,59	<0,01	2,64	0,16	0,29	0,53	528	50	40
V	µg/L	ICP-MS	0,01	2,2	1,42	2,3	<0,01	11,98	0,8	1,42	2,92	2396		
Zn	µg/L	ICP-MS	0,01	17,09	1,5	40,4	<0,01	207,1	0,4	1,5	12,4	41420		

Table 1: An overview of general statistics of parameters determined in the analyzed bottled waters (n=36) together with guideline values established by Iranian legislation for drinking water (ISIRI, 1997) and by the WHO (2017).

region. Because of an uneven distribution of annual precipitation, the number and quality of commercially exploited aquifers vary across the country. Accordingly, the majority of samples were taken from the Zagros and Alborz units.

2.2. Bottled water preparation

Among a large number of different brands of mineral bottled water in Iran, 36 of the well-known and top-selling brands were purchased from randomly selected local supermarkets from all over the country. The selected samples were gas-free mineral waters of the same production date and were stored in 0.5 l polyethylene terephthalate (PET) plastic bottles. The bottled mineral waters were purchased in duplicate

for quality control purposes and stored in the refrigerator prior to analysis.

2.3. Bottled water analysis

The purchased bottled mineral water samples were analysed at the Federal Institute for Geosciences and Natural Resources (BGR) in Hannover, Germany with the procedures described by Birke et al. (2010a,b), and Reimann and Birke (2010). Chemical elements, ions, and parameters were analysed using the following techniques: inductively coupled plasma optical emission spectrometry for major elements of Ca, Mg, K, Na, PO₄, SiO₂ and Sr; ion chromatography for major anions and cations of Br, Cl, F, NO₂, NO₃ and SO₄; inductively coupled plasma mass spectrom-

etry for 54 trace elements; titration method for HCO_3^- ; photometric method for NH_4^+ ; potentiometric method for pH, and conductometric method for electrical conductivity (EC).

3. Results

3.1. Physical parameters and major elements

The pH values of bottled mineral water vary from 7.27 to 8.12, with a median of 7.81, which fall in the range of near neutral to slightly alkaline water. The median value of EC of all samples is $347 \mu\text{S}/\text{cm}$ (ranging from 95 to $742 \mu\text{S}/\text{cm}$). Total dissolved solids (TDS) varies between 62 and 482 mg/l (median: 226 mg/l). Total hardness (TH) varies from 33 to 325 mg/l, with a median of 162 mg/l.

Bicarbonate is dominant in 30 of the bottled water samples with the median and maximum values of 172 mg/l and 322 mg/l, respectively (Tab. 1). Zagros bottled mineral samples have the highest bicarbonate concentration at 322 mg/l. The content of sulphate in the mineral water samples is generally low except for four brands with SO_4 higher than 40 mg/l. The analysed bottled mineral water samples are characterized by low concentrations of chloride (median: 7.60 mg/l) and fluoride (median: 0.128 mg/l). The highest chloride value (59.4 mg/l) was measured in one brand from Kerman province, Central Iran. Bottled mineral water samples contain various levels of nitrate ranging from 0.16 to 36.2 mg/l. Nitrite yielded values below 0.01 mg/l. Ammonium ion and ammonia were not detected in the analysed bottled mineral water samples. Calcium (median: 46.2 mg/l), sodium (median: 8.6 mg/l) and potassium (median: 0.65 mg/l) are present in concentrations that progressively decrease in the order listed. Magnesium has a wide concentration range, varying from 0.04 to 36.5 mg/l with a median of 9.89 mg/l.

3.2. Trace elements

The median value for most trace elements (As, Cd, Co, Cs, Cu, Fe, Hg, Ni, Pb, Rb, Sb, Sc, Se, Zr) is less than $0.5 \mu\text{g}/\text{l}$ in all of the studied bottled mineral water samples (Tab. 1). The elements Ag, Be, Bi, Br, Ce, Dy, Er, Eu, Ga, Gd, Ge, Hf, Ho, La, Lu, Nb, Nd, Pr, Sm, Ta, Tb, Te, Th, Ti, Tl, Tm, W, Y and Yb occur in very low concentrations and mostly below their detection limits. Bottled mineral water yielded concentrations of Al (0.06 to $5.51 \mu\text{g}/\text{l}$), Fe (0.04 to $1.56 \mu\text{g}/\text{l}$) and Mn (0.01 to $0.54 \mu\text{g}/\text{l}$). The greatest variability (maximum value divided by minimum value) was observed for As, Ba, Cr, Cu, I, Li, Mo, Se, V and Zn where the

concentrations differ by several orders of magnitude. The concentration of As is significantly high ($58 \mu\text{g}/\text{l}$) in one sample collected from Kerman province, while the amount of As in the other bottled mineral water samples vary between 0.01 to $1 \mu\text{g}/\text{l}$. The maximum permissible value of As is $10 \mu\text{g}/\text{l}$ (WHO, 2017; ISIRI, 1997). Furthermore, this sample together with one from Zanzan contains considerable amounts of boron at 522 $\mu\text{g}/\text{l}$ and 692 $\mu\text{g}/\text{l}$, respectively. Concentration of boron in the other samples range between 4 and 132 $\mu\text{g}/\text{l}$. The WHO standard (2017) suggests a permissible value of 2400 $\mu\text{g}/\text{l}$ for boron.

4. Discussion

The pH value range in bottled mineral water samples is in accordance with the statutory range of Iranian standards for potable water (pH 6.5 to 9.0). All bottled mineral water samples are within the optimal European Commission limits for bottled mineral water (Birke et al., 2010b), and thus reflect adequate content in TDS.

Concentration of most major ions complies with recommendations by ISIRI (1997) and WHO (2017). Based on piper diagrams, thirty out of the 36 analysed bottled mineral water samples are of Ca-HCO_3 hydrogeochemical type. No health-based guideline is proposed for HCO_3^- . The taste limit for calcium is between 100 and 300 mg/l (Van der Aa, 2003). Hydrochemical types of bottled mineral water samples do not change over the four structural units of Iran, but presents a relatively broader variation in samples drawn from different subdivisions of the Alborz unit. Five of the analysed bottled mineral waters, from Alborz, Ardebil, Tehran (two samples) and Zanzan provinces, are of Na-HCO_3 , Ca-SO_4 , Ca-Cl , Mg-SO_4 and Na-SO_4 types, respectively.

Concentration of sulphate in samples is relatively low (median: 14.60 mg/l) and within the safe standard limit of 400 mg/l proposed by ISIRI (1997). Regarding NO_3^- , concentrations of up to 9 mg/l could be considered to be naturally occurring in groundwater (WHO, 2017). Samples with higher contents of NO_3^- can be related to the influence of agricultural fertilizers. Chlorine and F⁻ contents are below the permissible values proposed by WHO (2017), i.e. 5 mg Cl/l and 1.5 mg F/l. Based on the Stuyfzand (1989) classification, 32 of the bottled mineral water samples from this study have a chloride content of less than 30 mg/l, and fall into the very oligohaline (< 5 mg/l Cl⁻) and oligohaline (5 to 30 mg/l Cl⁻) classes. Based on the European Commission classification for bottled mineral water types, as discussed by Birke et al. (2010b), none of the studied samples contain values in excess, i.e.

$\text{HCO}_3^- > 600 \text{ mg/l}$, $\text{SO}_4^{2-} > 200 \text{ mg/l}$, $\text{Cl}^- > 200 \text{ mg/l}$, $\text{Ca}^{2+} > 150 \text{ mg/l}$, $\text{Mg}^{2+} > 50 \text{ mg/l}$, $\text{F}^- > 1 \text{ mg/l}$, and $\text{Na}^+ > 200 \text{ mg/l}$). Due to the very low sodium content of the bottled mineral water samples they mostly fit into the "suitable for low sodium diets" ($\text{Na}^+ < 20 \text{ mg/l}$).

Trace elements have very low concentrations in bottled mineral water samples and comply with the limits set by national and international standards. Nevertheless, the amount of arsenic and boron is remarkably high in one brand of bottled mineral water sourced in the Central Iran lithological zone. Further research is recommended to elaborate the specific lithological factors causing this anomaly.

The difference between the elemental contents of the analysed bottled mineral water samples, based on their origin in variety of geological and structural units of Iran was verified. The null hypothesis for this test is that the natural concentrations of elements in water samples are not influenced by local geology. This hypothesis rejects when the attained significance level (p) is less than or equal to 0.05. The Kruskal-Wallis test revealed significant differences for boron ($p\text{-value} = 0.028$), nickel ($p\text{-value} = 0.017$), potassium ($p\text{-value} = 0.031$), strontium ($p\text{-value} = 0.008$), bromine ($p\text{-value} = 0.050$) and sulphate ($p\text{-value} = 0.016$). The rest of the components showed no difference in various lithological units ($p\text{-value} > 0.05$).

In general, cations cannot be regarded as an impression of the impact of regional geology because the essential cations of Ca and Mg are originated from both, calcareous and volcanic rocks. Anions are better indicators for revealing such correlation. Samples taken from Zagros contain high bicarbonate concentrations indicating karstic spring sources. Elevated amounts of sulphate in four of the samples can be attributed to: i) the black shale of the Jurassic Shemshak Formation underlying the source rock limestone in an Alborz sample; ii) the marl and evaporate units of the Upper Red Formation underlying the alluvium in Zanjan sample in Alborz Zone; iii) impact of the evaporate unit beneath the volcanic rocks in Kopet-Dagh; iv) the evaporate units underlying the limestone in Central Iran sample.

5. Conclusion

The bottled mineral water samples used in this study represent the top 36 selling brands in Iran. The quality of these water samples generally complies with Iranian (ISIRI, 1997) and WHO (2017) standards for drinking water. Nevertheless, one brand seems not to be suitable for consumption due to containing exceeding amounts of arsenic. This brand of bottled mineral water has its source in Oligo-Miocene car-

bonates of Central Iran unit. However, we are not certain which lithological factors are responsible for this anomaly. Chemical differences between the bottled mineral water samples in four Iranian geological units are mainly reflected in the content of anions such as carbonate and sulphate. In addition to water-rock interactions, climatic parameters may have contributed to the contents of the studied bottled mineral water samples.

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