

Chemical composition and heavy metals in bottled mineral drinking waters in Bandar Abbas

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Abstract

Introduction: In recent years, attraction toward drinking bottled mineral water has increased. Therefore, chemical composition and safety of these waters considering trace elements and heavy metals should confirm the national or international standard limits. The aim of this study was to evaluate and comparison of the mineral and heavy metal composition of bottled waters in Bandar Abbas with Iranian national standard range.

Methods: A total of 125 samples from 25 bottled mineral water brands were analyzed for 17 elements including Zn, Cu, Ca, Mg, Na, K, Al, Mn, As, Pb, Cr, Se, Cd, Co, Ni, Mo, Ba using ICP-OES technique. Concentrations were compared to the desirable and permissible limits of Iran national standard.

Results: Zn, Cu, Ca and Mg were lower than desirable limits. Na concentration in all brands was lower than maximum permissible limit. Concentrations of Cr and Se were higher than maximum permissible limit in 7 brands. As, Pb, Cd, Co, Ni, Mo and Ba concentrations were lower than maximum permissible limit in all brands.

Conclusion: The results showed that the studied mineral waters could not provide clinically important dietary intake of Ca, Mg, Cu and Zn. Low Na concentration was a proper characteristic. Except for 7 brands with high Cr and Se concentrations, other bottled mineral waters were safe for daily intake.

Key words: Bottled Water, Heavy Metal, Trace Element

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Introduction:

Bottled mineral waters containing various dissolved minerals are now widely used. In the world, different kinds of bottled waters like spring, purified or sparkling bottled waters are used. According to the International Association of mineral water definition, waters with at least 250 ppm total dissolved solids are categorized as

mineral water (1). Natural mineral waters usually originate from underground deposit and do not need purification process (2). Also the chemical composition of natural mineral water is constant (3). In some areas with low quality water supplies bottled mineral water is usually used (4,5).

Historically, mineral waters were used for medical aims (6). The chemical composition of mineral waters depends on the geological area.

Since the amounts of trace elements such as arsenic or chromium and concentration of minerals (Ca, Mg, Zn, ...) are different in mineral waters, then using different mineral waters have different health outcomes. Epidemiological studies revealed that chemical composition of drinking water is related to certain forms of cancers (7), some congenital malformations (8) and even cardiovascular disease (9). On the other hand, minerals such as Ca can increase the mineralization of bone compensate the deficiency of this mineral in diet (10,11). Also Ca rich mineral waters are very useful for normal growth of children (12). Patients suffering from gout and hyperuricemia are recommended to use natural mineral waters (13).

One of the other properties of mineral waters is acid-base properties. The acid-base equilibrium of the body is very crucial for health and even very small changes of pH can widely change the cellular functions (14). In Iran, there were different mineral water resources and study on the properties of these waters started in the past decade (15). Especially in south of Iran, quality of natural water resources is not appropriate and bottled mineral waters are very popular. The consumption of tap water has declined and mineral water has attracted more interest. In 2008, Rezaee et.al studied 6 chemical parameters of 16 brands of mineral water in Yasouj and reported that the measured parameters were at the standard level (16). Another study in Hamedan (2014) evaluated only pH, alkalinity, nitrate, sulfate, phosphate, sodium and potassium and reported that the concentrations of these chemical were in standard range (17). In Qom nitrate and nitrite content of 18 bottled drinking waters were studied and the results showed that in 33% of samples, nitrate was higher than standard level (15). Another study on 13 bottled mineral water brands in Kerman revealed that concentration of K and Na were higher than permitted level in 23% and 46% of the samples respectively (18). In only one study, Hadiani et.al investigated 5 heavy metals in bottled waters and reported that the values were in permissible limit (19). With respect to the widespread usage of bottled waters in Bandar Abbas and the importance of chemical composition of drinking waters on human health, this study was aimed to determine the chemical composition of bottled waters in Bandar Abbas and comparing

them to the Iranian national standard values. Finally an overview of the chemical composition of the mineral waters will be discussed.

Methods:

In this cross-sectional study, a total of 125 samples were collected from 25 bottled water brands (5 samples per brand). Only 1 brand was produced in Hormozgan province and other brands were produced in other provinces. All brands were packed in polyethylene terephthalate (PET) containers with plastic screw caps. Analysis was performed in Central laboratory of Hormozgan University, Bandar Abbas, Iran.

All dishes and vials were cleaned prior to use with 5% HNO₃ and rinsed with MilliQ1 water. 50 mL of samples were transferred to falcon tubes and degased. 5 mL of all samples were treated with ultra-pure nitric acid 1% (v/v). Samples incubated at room temperature for 1 hour. All samples were filtered prior to analysis.

Standard solutions of all determined elements (1000 mg L⁻¹) were purchased from sigma (USA).

Calibration curve of Na, Mg and Ca were prepared using 0.5-1000 mg L⁻¹ of standard solution. The calibration range of Mn, Al, K, Cu, Zn, Ba, Mo, Ni, Co, Cd, Se, Cr, Pb and As were 5μg L⁻¹ to 100 mg L⁻¹. Ultra-pure HNO₃ were purchased from sigma. pH was measured using a metrohm pH meter (CH-9100 Herisau, Switzerland).

ICP-OES measurement

17 elements were analyzed using ICP-OES (725-ES, Agilent Technologies Inc.). Analytes were simultaneously analyzed by a slurry nebulizer and a charge coupled device detector. A high-accuracy nebulizer by free aspiration and a gravity drain was used to introduce samples into the plasma. Argon was used as the polychromator purge gas. The following emission lines were selected for each element according to their sensitivity: Na (588.8 nm), Ca (317.9 nm), K (769.9 nm), Mg (285.2) nm, Pb (283.3 nm), Ni (231.6 nm), Mo (284.8 nm), Mn (293.3 nm), Cu (223 nm), Cr (206.5 nm), Co (238.9 nm), Cd (326.1 nm), Ba (614.2 nm), As (228.8 nm), Al

(237.3 nm), Zn (334.5 nm), Se (203.98 nm). Operating condition for analysis of elements was shown in Table 1.

Table 1. Operating conditions employed in ICP OES spectrometer (Agilent 725-ICP-OES) for the analysis of water samples

Parameter and accessories	Value
Power (kw)	1.2
Plasma gas flow rate (L/min)	15
Auxiliary gas flow rate (L/min)	1.5
Nebulizer flow (L/min)	0.75
viewing height (mm)	10
Replicate read time (s)	1
Instrument stabilization delay (s)	15
Sample uptake delay (s)	30
Pump rate (rpm)	15
Exposure time (s)	15
Rinse time (s)	30
Plasma view	Vertical torch; axial/radial view
Background correction	2 points
Gas type	Argon
Number of replicates	3

After fully calibration of the instrument, nitric acid blank signal was measured in 3 replicates and the concentration equal to three times the standard deviation of this signal measurement considered as limit of detection (LOD). The plasma flow rate was 15 L min⁻¹. The linear range of calibration curves for all elements were accurately determined.

Statistical Analysis

SPSS statistical software package (version 19.0; SPSS Inc., Chicago, IL, USA) was used for data analysis. Mean, median and standard deviation of the variables were presented. Finally the elements concentrations were compared with Iranian national standard range.

Results:

Mineral water chemistry

In this study the mineral composition of the bottled waters were measured and compared to the Iran national standard (1053) range (20,21). As is shown in Table 2, Zn (mean±SD 1±0.29 ppm, median 0.9 ppm, min 0.7 ppm, max 1.76 ppm), Ca (mean±SD 43.2±18.4 ppm, median 44.2 ppm, min 3.4 ppm, max 75.6 ppm) and Mg (mean±SD

8.4±5.1 ppm, median 8.4 ppm, min 0.19 ppm, max 17.6 ppm) contents of all 25 brands were significantly lower than desirable Limit. Na concentration of all 25 brands were lower than 200 ppm (mean±SD 16.3±19.98 ppm, median 6.3 ppm, min 1.03 ppm, max 82.4 ppm) and hence, Na concentration was in standard range. The concentrations of K (mean±SD 0.98±0.9 ppm, median 0.5 ppm, min 0.11 ppm, max 3.2 ppm), Cu (mean±SD 0.038±0.02 ppm, median 0.038 ppm, min 0.003, max 0.08 ppm) and Mn (mean±SD 0.02±0.028 ppm, median 0.01 ppm, min <DL ppm, max 0.08 ppm) in mineral waters were lower than the standard desirable limit. Al concentrations (mean±SD 0.04±0.036 ppm, median 0.03 ppm, min <DL ppm, max 0.17 ppm) were higher than desirable limit in 1 brand and lower than desirable limit in other brands.

Mineral waters toxic elements

Concentrations of 9 toxic elements that were measured in 25 bottled mineral waters were shown in Table 3. The concentration of these elements were compared to the national permissible limit of Iran (20,21). Considering Pb, except for three brands, in all other brands, concentration of Pb was lower than maximum permissible limit (mean±SD 0.008±0.002 ppm, median 0.007 ppm, min <DL ppm, max 0.013ppm). For the three brands (brands number 1,11 and 12) Pb concentration was around maximum permissible limit. Chromium was another toxic element that was measured in this study. The mean concentration of Cr was 0.045±0.02 ppm (median 0.04 ppm, min <DL ppm, max 0.088 ppm). From 25 bottled mineral water, 9 brands showed Cr concentration below detection limit. Cr concentration was higher than permissive limit (0.05 ppm) in 3 brands. Ba, Mo, Ni, Co and As concentrations in all mineral water brands were lower than permissive limit (Table 3).

Mean concentration of these toxic elements were as follows: Ba, 0.041±0.026 ppm (median 0.049 ppm, min <DL ppm, max 0.074 ppm), Mo, 0.046±0.02 ppm (median 0.053 ppm, min 0.01 ppm, max 0.071 ppm), Ni, 0.048±0.019 ppm (median 0.056 ppm, min 0.012 ppm, max 0.071 ppm), Co, 0.024±0.01 ppm (median 0.026 ppm, min <DL ppm, max 0.054 ppm), As, 0.006±0.001 ppm (median 0.066 ppm, min <DL

ppm, max 0.0096 ppm). Selenium (Se) and Cadmium (Cd) concentrations were also measured in mineral waters produced by 25 Iranian companies. Mean concentration of Se was 0.0077 ± 0.004 ppm (median 0.006 ppm, min <DL, max 0.017 ppm) and the mean concentration of Cd was 0.0018 ± 0.004 ppm

(median 0.0018 ppm, min 0.001 ppm, max 0.003 ppm). Se concentration in 3 mineral water brands was higher than standard maximum permissible limit. Cd concentration in all mineral water brands was lower than maximum permissible limit.

Table 2. Permissible and desirable limit of minerals according to the Iran national standard (1053) and concentrations of the same minerals in 25 brands of bottled mineral waters

	Mn (ppm)	Al (ppm)	K (ppm)	Na (ppm)	Mg (ppm)	Ca (ppm)	Cu (ppm)	Zn (ppm)	pH
Desirable limit	0.1	0.1	5	-	30	300	1	3	6.5-8.5
Maximum permissible limit	0.4	0.1-0.2	50	200	-	-	2	-	6.5-9
1	0.015	0.013	0.506	22.06	10.7343	44.2	0.013	1.168	7.3
2	0.012	0.051	0.515	5.19	7.02476	66.5	0.028	0.747	7.5
3	0.047	0.012	0.433	1.03	5.85511	40.9	0.082	1.003	7.3
4	0.014	0.026	0.517	2.55	1.52697	69.2	0.013	0.770	8
5	< DL*	0.036	3.033	13.78	13.3461	75.6	0.061	0.792	7.4
6	0.058	0.032	0.747	23.76	4.9791	34.6	0.054	1.100	7.2
7	< DL	0.020	0.528	1.74	12.3075	51.0	0.038	0.903	7.4
8	0.058	0.031	0.306	22.54	11.0083	43.3	0.040	1.482	7.2
9	< DL	0.005	0.309	9.83	12.5242	53.3	0.032	0.705	7.3
10	< DL	0.079	0.229	1.17	4.58042	53.0	0.062	0.990	7.4
11	0.047	0.048	3.229	63.46	16.3521	44.7	0.056	1.234	7.3
12	0.022	0.098	0.454	6.27	15.6577	48.0	0.047	0.817	7.5
13	< DL	0.020	2.785	3.91	8.40868	69.5	0.039	0.902	7.3
14	< DL	0.036	0.512	21.57	10.5432	44.2	0.025	0.729	7.6
15	< DL	0.018	0.210	4.81	0.194274	42.6	0.038	0.736	7
16	0.061	< DL	0.206	21.94	9.17559	48.3	0.032	0.831	7.4
17	0.070	0.048	1.770	35.27	1.45465	6.2	0.037	1.758	7.7
18	0.040	0.001	0.508	1.87	1.41881	3.4	0.042	0.830	7.5
19	< DL	0.025	0.979	3.46	17.6112	40.3	0.022	0.788	7.6
20	< DL	0.054	0.986	22.66	9.26057	42.6	0.019	0.947	7.6
21	0.034	0.174	0.106	1.75	5.05715	49.4	0.063	0.924	7.5
22	0.082	0.039	0.975	26.17	15.4323	41.1	0.003	1.393	7.5
23	0.069	0.060	1.880	82.42	6.41935	36.1	0.038	1.689	7.5
24	0.011	0.054	0.953	2.59	3.4928	27.6	0.025	0.920	7.9
25	< DL	0.016	1.838	5.99	5.3328	4.9	0.054	0.941	7.1
Detection limit ($\mu\text{g/L}$)	0.05	0.2	2	0.5	0.07	0.3	0.2	0.3	-

* DL detection limit

Table 3. Maximum permissible limit according to the Iran national standard (1053) and concentration of toxic elements in bottled mineral waters

	Ba (ppm)	Mo (ppm)	Ni (ppm)	Co (ppm)	Cd (ppm)	Se (ppm)	Cr (ppm)	Pb (ppm)	As (ppm)
Desirable limit	-	-	-	-	-	-	-	-	-
Maximum permissible limit	0.07	0.07	0.07	0.05	0.003	0.01	0.05	0.01	0.01
1	.06611	.06800	.07000	.01800	.0020	.00549	<DL	.0114	.0066
2	.03600	.06100	.07000	.03700	.0031	.00542	<DL	.0086	.0050
3	<DL	.05700	.05900	.03000	.0015	.00244	<DL	.0073	.0090
4	.01200	.07000	.06300	.03700	.0017	.00902	.0290	.0076	.0045
5	.06502	.07111	.05900	.01700	.0012	.00632	.0711	.0061	<DL
6	.06812	.05400	.01590	.01200	.0022	.00927	<DL	.0064	.0096
7	.00700	.06000	.04700	.03000	.0017	.00688	.0570	.0083	.0069
8	.01090	.04500	.01340	.01700	.0025	.01400	.0370	.0049	.0062
9	.06200	.05000	.04400	.02600	.0014	.00768	.0441	<DL	.0032
10	.00400	.02800	.05900	<DL	.0013	.00534	<DL	.0087	.0044
11	.07380	.06300	.02400	.03500	.0014	.00561	.0360	.0126	.0060
12	.04900	.01460	.06600	.01800	.0020	.00493	.0390	.0135	.0082
13	.02200	.02900	.06010	.01500	.0021	.00513	.0400	.0073	.0041
14	.06700	.05300	.03200	.01500	.0018	.00633	<DL	.0061	<DL
15	.01300	.05900	.05000	.00000	.0015	.00624	.0511	.0068	.0030
16	.01280	.01160	.05500	.02700	.0023	.00583	.0440	<DL	.0072
17	.01530	.04400	.03400	.01600	.0025	.00822	.0030	.0058	.0060
18	<DL	.04000	.06106	.02300	.0021	.01600	.0360	.0045	.0077
19	.06400	.01010	.05600	.02700	.0010	.01620	.0460	<DL	.0070
20	.06114	.04300	.06070	.01200	.0017	.00494	.0350	.0072	.0070
21	<DL	.06400	.01220	<DL	.0012	.00417	<DL	.0067	.0070
22	.06412	.01110	.01290	.04400	.0018	.01720	<DL	.0087	.0090
23	.07037	.06400	.06800	.02800	.0019	<DL	.0880	.0055	.0080
24	<DL	.01100	.06500	<DL	.0014	.00704	<DL	.0092	.0042
25	.02800	.06200	.04400	.02600	.0018	.00420	.0716	<DL	.0056
Detection limit ($\mu\text{g/L}$)	0.3	0.2	0.3	0.2	0.1	1.1	0.2	0.5	1.2

Conclusion:

In accordance with our results, Rezaei et.al. measured K, Na, Ca and Mg in 16 bottled mineral waters and reported that in all brands the concentrations of these minerals were lower than standard limits (16). Study of the mineral composition of the bottled water in North America revealed that these waters contained high levels of Ca and Mg (22). Another study of European bottled mineral waters used ICP-Mass method and showed that the samples contained high amounts of Ca and Mg and Zn (23). These cations are clinically significant for human health. Enzymes such as carbonic anhydrase use Zn as cofactor (1).

Ca and Mg are essential ions for bone mineralization and any disturbance in Ca homeostasis may cause serious health problems (24). Adults need at least 1000 ppm Ca per year

(22). Hence, drinking waters could provide parts of mineral needs of the body. Ca bioavailability of mineral waters is similar to dairy products (12) and could decrease bone resorption (25). Na is another important mineral in the body which is responsible for key processes such as neurotransmission.

Other mineral ions including K, Cu, Al and Mn are trace elements that play role in body function. K is important in water and electrolyte balance in the body (26). Mn is required for normal nerve functions and cellular homeostasis (27). Little amounts of Al can reinforce the immune response (28). Cu is also essential for enzyme functions and plays role in metabolic functions (29,30).

In one study by Riahi et.al., K and Na in seven brands of bottled mineral water were measured in Hamedan and they found that these minerals concentrations were in standard range. They used flame photometer instrument for the measurements

(17). Study of the bottled mineral waters in Kerman revealed that K concentration was higher than standard limit in 23% of the samples and 46% of the samples contained Na higher than standard limit (18). However the differences in the results could be attributed to different methods used for the analysis (flame photometer instead of ICP-OES) and also different brands of mineral waters. Mn was measured in European bottled mineral waters and its concentration was higher than the standard limit (23). These differences may be attributed to the different geochemical conditions in different area.

Pb is one of the environmental contaminations which are dangerous for human health (31). Using waters that contain lead is one way for entry of this element to the human food chain. Recycling old lead products and uses of leaded gasoline are responsible for environmental contamination and this contamination may pollute the water resources (31). Study of bottled mineral water in Italy showed that the concentration of Pb was higher than standard (32). Another study in Pakistan revealed that some bottled mineral waters contained high amounts of Pb (33).

Hadiani et al. studied bottled mineral water collected randomly from all parts of Iran. They showed that Pb concentration in all mineral waters were lower than standard range (19). The results of their study were in accordance with our results.

In human, trace amounts of Co are required for enzyme functions (34). Ba, Mo, Ni and As are dangerous for human health and their adverse effect on cardiovascular system (35,36), reproductive organs (37) and nervous, respiratory, immune and endocrine systems have been reported (38).

Therefore, the concentration of these elements in drinking mineral or ground water should not exceed the standard maximum permissible limit. Hadiani *et.al* measured As in 22 bottled mineral water in Iran. They found that the concentration of As in 6 samples exceeded the national maximum permissible limit of Iran (19). In another study, fifty six bottled mineral water were randomly gathered from all parts of Europe and analyzed. Ba and As were higher than standard limits in 16.1% and 8.9% respectively. In accordance with this study, Mo, Ni and Co concentration were lower than standard (23). Another study of bottled mineral

water from 23 European countries revealed that 2.8% of samples did not confirmed the European limits for Ba and Ni (39). Nazemi et al. measured the concentration of Se in waters from north, center and south of Iran. Se concentration was lower than detection limit. However they analyzed water from agricultural fields not the bottled mineral water (40). In accordance with this study, Hadiani et al. reported that Cd concentration of 42 bottled mineral water in Iran were permissible according to the limits of national and international standards (19).

Another study from drinking waters in Malaysia measured heavy metals containing Cd, Se, Cr, As and Hg. The concentrations of these elements compared to National Drinking Water Quality Standard of Malaysia and all were within safe limits (41). Miranzadeh et al. measured chromium, cadmium, lead, nickel, copper, zinc, and silver in 15 brands of bottled drinking waters in Iran and reported that concentration of these heavy metals were in range of Iranian standard values (42).

In this study, the chemical analysis of bottled mineral waters was conducted and the safety of the waters was evaluated. Concentrations of 17 elements were measured using ICP-OES technique. The strength of this study is that for the first time the concentration of 17 mineral and toxic elements were measured in Iranian bottled mineral waters. Our results showed that Ca, Mg, Zn and K concentrations were below the desirable limit. These elements have beneficial effect on human health. Our results showed that the 25 studied brands had little nutrition value regarding these minerals. On the other hand 7 mineral water brand (28% of the samples) had Cr and Se higher than maximum permissible limit. Hence these mineral waters may not be appropriate for human consumption and need more investigation to find the reason for this contamination. Anyway some limitations of this study should be mentioned: First, we analyzed the bottled mineral water gathered from Bandar Abbas and hence the variation of minerals in bottled water from all parts of Iran may be different from our results. Second, we did not measure fluoride, nitrate and nitrite and these ions are associated with benefits and risks for public health.

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ترکیب شیمیایی و فلزات سنگین آب‌های معدنی بطری شده در سطح شهر بندرعباس

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چکیده

مقدمه: در سال‌های اخیر، تمایل برای استفاده از آب‌های معدنی بطری شده افزایش یافته است. بنابراین ترکیب شیمیایی و سلامت این آب‌ها به خصوص از نظر فلزات سنگین و عناصر کمیاب باید با استانداردهای ملی و یا بین‌المللی مطابقت داشته باشد. هدف از این مطالعه، اندازه‌گیری فلزات سنگین و موثد معدنی موجود در آب‌های معدنی بطری شده در سطح شهر بندرعباس و مقایسه آن با استاندارد ملی آب‌های ایران بود.

روش کار: در این مطالعه، ۱۷ عنصر شامل *Zn, Cu, Ca, Mg, Na, K, Al, Mn, As, Pb, Cr, Se, Cd, Co, Ni, Mo, Ba* در ۱۲۵ نمونه آب معدنی از ۲۵ برند مختلف با استفاده از تکنیک *ICP-OES* اندازه‌گیری شد. غلظت عناصر مذکور با مقادیر مطلوب و مجاز ذکر شده در استاندارد ملی آب‌های ایران مقایسه گردید.

نتایج: مقادیر عناصر *Zn, Cu, Ca, Mg* از میزان مطلوب ذکر شده در استاندارد ملی آب‌های معدنی کمتر بود. غلظت *Na* در همه نمونه‌های بررسی شده از حداکثر مجاز کمتر بود. غلظت *Cr* و *Se* در ۷ برند آب معدنی نسبت به حداکثر مجاز بیشتر بود. غلظت عناصر *As, Pb, Cd, Co, Ni, Mo, Ba* در همه برندهای مورد بررسی کمتر از حداکثر مجاز ذکر شده در استاندارد ملی آب‌های معدنی بود.

نتیجه‌گیری: نتایج این مطالعه نشان داد که هیچ یک از آب‌های معدنی مورد بررسی، دارای غلظت مناسبی از عناصر *Ca, Mg, Cu, Zn* از نظر تأمین روزانه این عناصر جهت حفظ سلامت انسان نمی‌باشند. در عین حال، غلظت پایین سدیم در نمونه‌های مورد بررسی ویژگی مثبت نمونه‌ها می‌باشد. به جز ۷ برندی که غلظت عناصر *Cr* و *Se* در آنها بالاتر از حد مجاز بود، دیگر نمونه‌های مورد بررسی از نظر مصرف روزانه سالم ارزیابی شدند.

کلیدواژه‌ها: آب‌های بطری شده، فلزات سنگین، عناصر کمیاب

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