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Determination of trace and heavy metals in bottled drinking water in Yemen by ICP-MS

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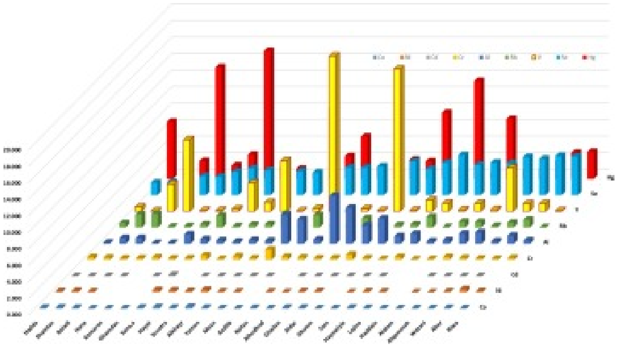
Abstract

This study assessed the concentrations of several trace and heavy metals in bottled drinking water. A total of 27 samples were collected from Yemeni markets. The ICP-MS (inductively coupled plasma-mass spectrometry) technique was utilized to analyze the levels of Li, Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Se, Rb, Ag, Cd, Cs, Ba, Hg, Tl, Pb, and U. Furthermore, the study also examined the levels of total dissolved solids (TDS), electrical conductivity (EC), and pH in the collected samples. The concentrations of the elements in the bottled drinking water samples were compared to the maximum allowable concentration (MAC) values for these elements in drinking water, set by the World Health Organization (WHO) and Yemen's Ministry of Water and Environment (YMWE). The limit of detection (LOD) and recovery ranged from 0.0003 to 1.86 µg/l, 80 to 120%, respectively. Only trace elements Mn and Pb were detected in all samples at concentrations below the LOD. Out of the elements examined, only Hg exceeded the acceptable limits defined by both the WHO and YMWE in most of the samples.

Graphical abstract



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Keywords

Bottled drinking water; Determination; ICP-MS; Trace Metals; Yemen

1. Introduction

Water is an essential resource for the survival of all living organisms, including humans. It is a natural element that reflects the geological features of the surrounding environment. Water can be found on the Earth's surface and underground. As it interacts with rocks and soil, it has the ability to dissolve various substances, both inorganic and organic. However, human activities, especially industrial processes, have significantly impacted the natural state of water. As a result, there is a growing recognition among individuals regarding the significance of preserving water cleanliness and assessing its suitability for drinking purposes [1].

The demand for bottled mineral water has grown significantly in response to concerns about water resources and water security. In recent years, more people have been choosing bottled mineral water over tap water. The increase in popularity can be attributed to urbanization, as it has led many individuals to prefer bottled water due to its affordability, convenience, lower contamination levels, and certified safety. While it is commonly believed that bottled water is safer than tap water, there have been cases where the composition of bottled water has exceeded standards or regulations for certain harmful natural chemicals and heavy metals [2], [3].

There is considerable apprehension surrounding the quality and safety of mineral waters in different communities. This is primarily due to the presence of various minerals and trace elements. Minerals play a vital role in supporting human health by providing nourishment, promoting growth, facilitating bodily functions, and contributing to overall well-being. However, the types and amounts of minerals can vary significantly depending on several factors.[4].

Marketing campaigns that highlight bottled water as a safe and refreshing alternative, along with discouraging the consumption of tap water, have played a significant role in boosting the sales of mineral water [5]. The desire for water that is perceived as both safe and enjoyable has been a key driving factor behind this trend [6], [7]. The belief in the medicinal and therapeutic benefits of natural bottled water has resulted in its preference over tap water, despite the existence of counterfeit mineral water in the market [6]. However, there are concerns about the quality of bottled water for various reasons, including the possibility of contamination during packaging, transportation, or storage [7], [8], [9].

Regulatory standards are established to define the acceptable elemental composition [3]. The human body needs a range of minerals, such as Ca, Mg, Co, Cu, Mo, Se, and Zn. However, there are also toxic elements like As, Cd, and Pb, as well as essential micronutrients, whose concentrations can become health hazards when they exceed certain thresholds. [10]. Nowadays, several studies have examined the elemental composition of bottled water in various countries, including Europe [3], Indonesia [11], Belarus [12], Greece [13], Croatia [14], Saudi Arabia [15], Turkey [4], and China [16]. In Yemen, the majority of Yemenis rely on untreated groundwater as their main source of drinking water [17], [18]. As a result, several researchers have undertaken studies to evaluate the elemental composition of groundwater and treated drinking water in Yemen [17], [19], [20].

The aim of this study was to examine the levels of trace and heavy metals and assess the physicochemical properties of bottled drinking water sold in the Yemeni markets.

2. Materials and methods

2.1. Sample collecting and preparation

A total of twenty-seven samples of bottled drinking water were collected from the Yemeni market in 2023. These samples were stored in a dark environment at temperatures between 15 °C and 20 °C and analyzed within three months of being bottled. The elements were analyzed without requiring sample dilution. Before measurement, all samples were acidified using 65% supra pure nitric acid (Fluka). The sample preparation was carried out using pre-cleaned polypropylene scintillation vessels.

2.2. Chemicals

Calibration solutions were prepared daily by diluting a 10 mg/L stock solution of an ICP multi-element standard solution, which included elements such as Li, Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Se, Rb, Ag, Cd, Cs, Ba, Tl, Pb, U, and Hg (from an Agilent Technologies, Palo Alto, CA, USA stock solution). The ICP-MS instrument calibration was performed using standard solutions ranging from 0.1 to 150 μg/L. Argon with a purity of 99.9999% (from White Martins, Saudi Arabia) was used as the plasma, nebulization, and auxiliary gas in the ICP-MS analysis.

2.3. Analytical methods

Purified ultrapure water with a conductivity of 0.055 μS/cm was obtained using a Barnstead water purification system ASTM Type II from Thermo Electron LED GmbH, Germany. The physiochemical parameters, including electrical conductivity, total dissolved solids (TDS), and pH, were measured using an Orion Star A215 pH/Conductivity Meter from Thermo Scientific, USA. The analysis of trace and heavy metals

(Li, Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Se, Rb, Ag, Cd, Cs, Ba, Hg, Tl, Pb, U) was conducted using ICP-MS 7800, Agilent, USA. The operating conditions of the ICP-MS are detailed in Table 1. All these measurements were performed at the central laboratory of the Chemistry Department, College of Science, Qassim University, Saudi Arabia.

Table 1. Operational conditions used for the determination of mineral elements by ICP-MS.

ICP-MS conditions	Value
RF generator power	1550
Plasma gas rate/(mL min-1)	30
Nebulizer type	MicroMist
Carrier gas (l/min)	5
Make up gas (l/min)	0.15
Plasma gas (l/min)	15
Nebulizer Pump (rps)	0.1
Points/peak	3
Repetitions	3
Integration time /mass (sec)	0.3

2.4. Analytical quality assurance

By utilizing calibration curves derived from standard solutions analyzed using ICP-MS, the concentrations of trace and heavy metals in acidified samples were assessed with three replicates to ensure accuracy. Several parameters, including the limit of detection (LOD) and accuracy (recovery level), were chosen to validate the method utilized in the study. The LOD was determined using the formula 3 times σ/S, where σ represents the residual standard deviation of the linear regression, and S denotes the slope of the calibration line (as shown in Table 4). To evaluate the accuracy of the method, recovery tests were performed by adding increasing amounts of each analyte to bottled drinking water samples at three spiking levels (10, 20, and 40 ppb). The results are presented in Table 2, and the mean recoveries for the elements ranged from 80% to 120%.

Table 2. Average recovery of elements from bottled drinking water samples.

Element/Concentration (ppb)	Li			Al			V			Cr			Mn	
Spike level	10	20	40	10	20	40	10	20	40	10	20	40	10	20
Mean result	10.11	20.76	46.95	11.67	21.59	47.85	9.86	23.30	47.78	9.85	21.26	46.53	9.89	23
Mean recovery (%)	101	104	117	117	108	120	99	117	119	99	106	116	99	11
Element/Concentration(ppb)	**Fe**			**Co**			**Ni**			**Cu**			**Zn**	

Element/Concentration (ppb)	Li			Al			V			Cr			Mn	
Spike level	10	20	40	10	20	40	10	20	40	10	20	40	10	20
Mean result	9.41	20.64	44.75	9.51	20.80	45.50	9.68	19.73	39.06	10.29	20.40	41.13	11.21	22
Mean recovery (%)	94	103	112	95	104	114	97	99	98	103	102	103	112	11
Element/Concentration (ppb)	**Ga**			**Se**			**Rb**			**Ag**			**Cd**	
Spike level	10	20	40	10	20	40	10	20	40	10	20	40	10	20
Mean result	9.60	21.71	44.378	9.92	21.51	42.35	10.23	23.05	47.65	8.68	18.68	43.67	10.04	20
Mean recovery (%)	96	109	111	99	108	106	102	115	119	87	93	109	100	10
Element/Concentration (ppb)	**Cs**			**Ba**			**Hg**			**Tl**			**Pb**	
Spike level	10	20	40	10	20	40	10	20	40	10	20	40	10	20
Mean result	9.95	20.51	41.92	11.56	23.11	43.27	11.54	21.49	44.30	9.52	19.54	47.62	10.46	19
Mean recovery (%)	100	103	105	116	116	108	115	107	111	95	98	119	105	97
Element/Concentration (ppb)	**U**													
Spike level	10	20	40											
Mean result	9.41	23.02	46.10											
Mean recovery (%)	94	115	115											

3. Results and discussion

3.1. Total dissolved solids (TDS), electrical conductivity (EC) and pH

TDS levels less than 600 mg/L are often regarded as having high palatability [21]. The WHO and YMWE have not established MAC values for TDS in drinking water[21], [22]. In contrast, the National Primary Drinking Water Guidelines by the US Environmental Protection Agency (US EPA) state that the total dissolved solids (TDS) level in drinking water should not exceed 600 mg/L[23]. The TDS levels of the examined bottled drinking waters ranged from 85.22 mg/L to 158.3 mg/L, with an average of 107.5 mg/L. These results are presented in Table 3.

Table 3. PH, total dissolved solids (TDS), and electrical Conductivity (EC) of investigated bottled drinking water brands.

samples	pH	TDS mg/L	EC µS/cm
Hadda	7.59	119.6	243.0
Shamlan	7.57	98.96	200.9
Beladi	7.72	125.7	254.2
Hana	7.87	110.7	224.9
Kamaran	7.58	101.6	206.2
Ghamdan	7.50	158.3	322.0
Sana·a	7.43	96.38	195.7
Hayat	7.35	110.6	224.7
Socotra	7.56	92.94	188.7
Alkhayr	7.43	123.5	251.0
Yemen	7.80	92.46	187.7
Mozn	7.85	111.3	226.1
Aadhb	8.20	84.14	170.7
Nofan	7.55	94.84	192.5
Alhodhod	7.72	112.2	228.0
Ghailan	7.81	113.1	229.7
Aldar	7.71	100.9	205.0
Ghuom	7.67	92.34	187.4
Sam	7.63	85.22	172.9
Hayawiyia	7.94	99.79	202.6
Lojina	7.68	89.35	181.3
Haddain	7.79	92.36	187.5
Alalam	7.74	121.6	247.2
Alqemmah	8.05	120.2	244.2
Watani	7.61	116.7	237.2
Abar	7.59	146.2	298.6
Riwa	7.52	92.42	187.6

There is no specific pH value recommended for health reasons. However, pH is regarded as one of the key indicators for evaluating water quality, even though its direct impact on consumers is uncommon [21]. The US EPA and YMWE recommend pH ranges of 6.5–8.5 and 6.5–9.0, respectively, However, the WHO does not provide specific guideline values for pH. The pH values of the tested bottled water samples in this study ranged from 7.35 to 8.20 (as shown in Table 2). These pH levels are considered acceptable based on both local and international regulations. EC is a measurement of ion concentration in water samples. In this study, the EC values of the examined bottled drinking water ranged from 170.7 to 322 µS/cm, with an average of 218.4 µS/cm. The average TDS and EC values for bottled drinking water samples in Yemen are comparable to those observed in bottled drinking waters produced in certain Arab countries [24].

3.2. Trace and heavy metals

3.2.1. Lithium

Lithium (Li) is known to be safe for human health. In recent years, there have been articles highlighting the positive effects of lithium on mental health. Furthermore, studies have indicated a connection between higher levels of Li in drinking water and a lower suicide rate. [25], [26]. The WHO and YMWE have not prescribed any MAC value for Li in drinking water[21], [22]. Table 4 shows that the levels of Li in the examined samples ranged from a minimum of 0.078 µg/L in the Riwa sample to a maximum of 18.751 µg/L in the Alalam sample. The average concentration of lithium in the studied samples is 2.041 µg/L.

Table 4. Concentration values (µg /L; mean±standard deviation) of the elements analyzed in Yemeni bottled waters.

Samples	Li	Al	V	Cr	Mn	Fe	Co
LOD	0.215	0.049	0.019	0.0075	0.036	0.131	0.004
WHO	–	–	–	50	–	–	–
YMWV	–	200	–	50	200	100	–
Hadda	0.753 ± 0.110	0.134 ± 0.019	0.510 ± 0.023	0.327 ± 0.019	< 0.036	0.204 ± 0.014	0.106 ± 0.006
Shamlan	1.521 ± 0.141	0.672 ± 0.077	0.051 ± 0.001	0.271 ± 0.024	< 0.036	< 0.131	0.132 ± 0.005
Beladi	0.988 ± 0.129	0.676 ± 0.052	3.266 ± 0.087	0.174 ± 0.011	< 0.036	< 0.131	0.087 ± 0.003
Hana	0.737 ± 0.102	0.081 ± 0.011	8.665 ± 0.201	0.186 ± 0.013	< 0.036	0.365 ± 0.021	0.009 ± 0.001
Kamaran	3.796 ± 0.434	0.093 ± 0.012	0.087 ± 0.014	0.310 ± 0.027	< 0.036	1.071 ± 0.136	0.010 ± 0.001
Ghamdan	2.933 ± 0.166	1.127 ± 0.019	0.100 ± 0.009	0.170 ± 0.012	< 0.036	0.523 ± 0.011	0.015 ± 0.001
Sana·a	1.584 ± 0.203	0.535 ± 0.050	0.271 ± 0.018	0.187 ± 0.033	< 0.036	< 0.131	0.112 ± 0.015
Hayat	0.314 ± 0.034	0.410 ± 0.014	3.433 ± 0.202	0.533 ± 0.048	< 0.036	< 0.131	0.181 ± 0.019

Socotra	0.580 ± 0.070	0.242 ± 0.016	1.078 ± 0.055	0.102 ± 0.010	< 0.036	0.910 ± 0.104	0.096 ± 0.011
Alkhayr	0.078 ± 0.011	0.454 ± 0.071	6.122 ± 0.844	0.442 ± 0.050	< 0.036	< 0.131	0.218 ± 0.028
Yemen	1.866 ± 0.239	0.366 ± 0.034	0.054 ± 0.010	0.177 ± 0.033	< 0.036	< 0.131	0.092 ± 0.010
Mozn	1.929 ± 0.281	3.455 ± 0.357	0.338 ± 0.052	1.263 ± 0.122	< 0.036	33.300 ± 0.100	0.030 ± 0.003
Aadhb	1.788 ± 0.215	2.941 ± 0.310	18.803 ± 1.376	0.383 ± 0.055	< 0.036	0.417 ± 0.040	0.012 ± 0.001
Nofan	1.239 ± 0.144	0.508 ± 0.051	0.051 ± 0.013	0.288 ± 0.031	< 0.036	< 0.131	0.070 ± 0.006
Alhodhod	1.631 ± 0.179	5.810 ± 0.666	0.305 ± 0.042	0.180 ± 0.024	< 0.036	7.265 ± 0.282	0.021 ± 0.003
Ghailan	2.023 ± 0.240	4.362 ± 0.629	0.076 ± 0.021	0.200 ± 0.031	< 0.036	< 0.131	0.096 ± 0.028
Aldar	0.471 ± 0.047	2.211 ± 0.185	17.240 ± 1.933	0.642 ± 0.089	< 0.036	5.949 ± 0.603	0.100 ± 0.013
Ghuom	1.741 ± 0.192	3.035 ± 0.281	0.113 ± 0.020	0.201 ± 0.022	< 0.036	0.711 ± 0.055	0.014 ± 0.002
Sam	0.769 ± 0.042	0.904 ± 0.133	1.352 ± 0.201	0.094 ± 0.010	< 0.036	< 0.131	0.029 ± 0.003
Hayawiyia	1.835 ± 0.159	1.224 ± 0.086	0.978 ± 0.129	0.355 ± 0.051	< 0.036	< 0.131	0.102 ± 0.008
Lojina	1.208 ± 0.054	0.293 ± 0.042	0.157 ± 0.022	0.054 ± 0.014	< 0.036	0.283 ± 0.011	0.066 ± 0.007
Haddain	1.114 ± 0.133	0.420 ± 0.059	0.960 ± 0.138	0.089 ± 0.012	< 0.036	< 0.131	0.021 ± 0.002
Alalam	18.751 ± 1.306	1.233 ± 0.110	0.194 ± 0.032	0.361 ± 0.039	< 0.036	7.313 ± 0.164	0.016 ± 0.002
Alqemmah	0.298 ± 0.022	1.388 ± 0.154	5.251 ± 0.840	0.233 ± 0.021	< 0.036	1.042 ± 0.107	0.029 ± 0.003
Watani	1.317 ± 0.120	0.301 ± 0.037	0.943 ± 0.141	0.150 ± 0.009	< 0.036	0.087 ± 0.010	0.034 ± 0.003
Abar	2.619 ± 0.181	0.972 ± 0.152	0.948 ± 0.110	0.132 ± 0.007	< 0.036	< 0.131	0.070 ± 0.005

Riwa	0.078 ±	0.008	0.404 ±	0.050	0.073 ±	0.014	0.227 ±	0.033	< 0.036	0.416 ±	0.041	0.030 ±	0.003	

Samples	Ni		Cu		Zn		Ga		Se		Rb		Ag	
LOD	0.012		0.006		1.860		0.0006		0.781		0.008		0.005	
WHO	70		2000		–		–		40		–		–	
YMWV	20		1000		15,000		–		10		–		–	
Hadda	0.128 ±	0.017	0.009 ±	0.001	< 1.860		0.028 ±	0.004	1.500 ±	0.140	0.508 ±	0.021	0.016 ±	0.00
Shamlan	0.171 ±	0.005	< 0.006		< 1.860		0.017 ±	0.001	1.572 ±	0.211	1.580 ±	0.068	<0.005	
Beladi	0.096 ±	0.005	0.096 ±	0.005	< 1.860		0.051 ±	0.006	2.654 ±	0.183	1.640 ±	0.082	<0.005	
Hana	< 0.012		0.173 ±	0.014	< 1.860		0.003 ±	0.001	2.301 ±	0.138	0.082 ±	0.007	<0.005	
Kamaran	< 0.012		0.016 ±	0.001	< 1.860		0.004 ±	0.001	2.217 ±	0.330	0.141 ±	0.008	<0.005	
Ghamdan	< 0.012		0.021 ±	0.001	< 1.860		0.020 ±	0.002	2.822 ±	0.385	0.376 ±	0.015	<0.005	
Sana,a	0.273 ±	0.016	< 0.006		< 1.860		0.009 ±	0.001	3.233 ±	0.452	1.434 ±	0.107	<0.005	
Hayat	0.222 ±	0.005	0.120 ±	0.005	< 1.860		0.004 ±	0.001	2.989 ±	0.294	0.134 ±	0.015	<0.005	
Socotra	0.140 ±	0.002	0.021 ±	0.002	< 1.860		0.008 ±	0.001	3.341 ±	0.474	0.318 ±	0.036	<0.005	
Alkhayr	0.299 ±	0.033	0.142 ±	0.011	< 1.860		0.004 ±	0.001	2.883 ±	0.178	0.376 ±	0.034	<0.005	
Yemen	0.080 ±	0.007	< 0.006		< 1.860		0.036 ±	0.004	2.744 ±	0.114	1.647 ±	0.132	<0.005	
Mozn	0.067 ±	0.027	0.692 ±	0.010	7.834 ±	0.702	0.198 ±	0.022	2.965 ±	0.173	0.363 ±	0.040	<0.005	
Aadhb	< 0.012		0.020 ±	0.001	< 1.860		0.138 ±	0.017	3.332 ±	0.403	1.502 ±	0.105	<0.005	
Nofan	0.070 ±	0.006	< 0.006		< 1.860		0.003 ±	0.003	3.322 ±	0.622	0.310 ±	0.025	<0.005	
Alhodhod	0.021 ±	0.001	< 0.006		< 1.860		0.162 ±	0.018	3.477 ±	0.081	0.322 ±	0.068	<0.005	

Ghailan	0.117 ±	0.015	< 0.006		6.968 ±	0.586	0.010 ±	0.001	3.884 ±	0.503	1.112 ±	0.133	<0.005	
Aldar	0.121 ±	0.014	0.066 ±	0.005	< 1.860		0.024 ±	0.002	4.080 ±	0.446	0.550 ±	0.066	<0.005	
Ghuom	< 0.012		0.051 ±	0.005	< 1.860		0.035 ±	0.003	3.167 ±	0.123	0.272 ±	0.038	<0.005	
Sam	0.011 ±	0.002	0.031 ±	0.002	< 1.860		0.006 ±	0.001	3.922 ±	0.345	0.355 ±	0.046	<0.005	
Hayawiyia	0.104 ±	0.015	0.063 ±	0.006	< 1.860		0.851 ±	0.046	4.828 ±	0.440	1.283 ±	0.115	0.076 ±	0.00
Lojina	0.085 ±	0.006	< 0.006		< 1.860		0.011 ±	0.002	3.655 ±	0.442	0.049 ±	0.004	0.023 ±	0.00
Haddain	< 0.012		0.010 ±	0.001	< 1.860		0.003 ±	0.001	3.921 ±	0.392	0.678 ±	0.075	0.013 ±	0.00
Alalam	0.006 ±	0.001	0.050 ±	0.003	< 1.860		0.037 ±	0.005	3.880 ±	0.150	0.650 ±	0.072	<0.005	
Alqemmah	0.008 ±	0.001	< 0.006		< 1.860		0.019 ±	0.003	4.600 ±	0.181	0.172 ±	0.014	0.012 ±	0.00
Watani	0.102 ±	0.006	0.315 ±	0.012	< 1.860		0.009 ±	0.001	4.382 ±	0.443	0.575 ±	0.052	<0.005	
Abar	0.387 ±	0.050	1.312 ±	0.015	3.213 ±	0.247	0.019 ±	0.002	4.745 ±	0.255	0.941 ±	0.085	<0.005	
Riwa	0.130 ±	0.018	< 0.006		< 1.860		0.003 ±	0.001	4.676 ±	0.481	0.024 ±	0.001	<0.005	
Samples	Cd		Cs		Ba		Hg		Tl		Pb	U		
LOD	0.0005		0.003		0.036		0.264		0.0017		0.0025	0.0003		
WHO	3		–		1300		6		–		10	10		
YMWV	5		–		–		1		–		5	–		
Hadda	0.0050 ±	0.0007	0.027 ±	0.002	18.066 ±	0.157	6.902 ±	0.338	0.0117 ±	0.0015	<0.0025	0.096 ±	0.013	
Shamlan	0.0023 ±	0.0003	0.016 ±	0.002	21.434 ±	0.122	2.888 ±	0.086	0.0062 ±	0.0008	<0.0025	0.009 ±	0.001	
Beladi	0.0062 ±	0.0007	0.088 ±	0.003	27.871 ±	0.334	2.159 ±	0.187	0.0053 ±	0.0008	<0.0025	0.015 ±	0.002	
Hana	0.0013 ±	0.0001	< 0.003		0.350 ±	0.020	13.506 ±	0.342	<0.0017		<0.0025	0.005 ±	0.001	

Kamaran	<0.0005		0.009 ±	0.001	0.630 ±	0.043	1.622 ±	0.233	0.0039 ±	0.0005	<0.0025	0.002 ±	0.000
Ghamdan	0.0091 ±	0.0014	0.010 ±	0.002	0.232 ±	0.014	2.910 ±	0.401	<0.0017		<0.0025	0.004 ±	0.001
Sana’a	0.1421 ±	0.0137	0.026 ±	0.004	20.328 ±	0.068	15.492 ±	0.242	0.0025 ±	0.0003	<0.0025	0.045 ±	0.004
Hayat	0.0007 ±	0.0001	0.006 ±	0.000	2.475 ±	0.029	1.465 ±	0.142	<0.0017		<0.0025	0.205 ±	0.025
Socotra	0.0016 ±	0.0002	< 0.003		1.701 ±	0.028	1.233 ±	0.154	<0.0017		<0.0025	0.019 ±	0.002
Alkhayr	0.0008 ±	0.0007	0.017 ±	0.002	3.534 ±	0.055	0.649 ±	0.066	0.0118 ±	0.0013	<0.0025	0.085 ±	0.008
Yemen	0.0032 ±	0.0004	0.016 ±	0.002	24.881 ±	0.331	8.612 ±	0.422	0.0023 ±	0.0003	<0.0025	0.009 ±	0.001
Mozn	0.0106 ±	0.0010	0.010 ±	0.001	0.718 ±	0.042	2.743 ±	0.224	<0.0017		<0.0025	0.004 ±	0.000
Aadhb	0.0022 ±	0.0002	0.016 ±	0.002	0.510 ±	0.044	5.133 ±	0.685	<0.0017		<0.0025	0.011 ±	0.001
Nofan	0.0026 ±	0.0001	0.016 ±	0.002	5.576 ±	0.010	1.487 ±	0.287	0.0140 ±	0.0010	<0.0025	0.040 ±	0.003
Alhodhod	0.0073 ±	0.0011	0.014 ±	0.002	0.152 ±	0.022	3.231 ±	0.223	<0.0017		<0.0025	0.004 ±	0.001
Ghailan	0.0016 ±	0.0001	0.033 ±	0.003	5.211 ±	0.047	2.321 ±	0.236	0.0113 ±	0.0017	<0.0025	0.007 ±	0.001
Aldar	0.0017 ±	0.0002	< 0.003		4.714 ±	0.018	2.124 ±	0.172	<0.0017		<0.0025	0.044 ±	0.007
Ghuom	0.0021 ±	0.0002	0.006 ±	0.001	0.440 ±	0.032	8.003 ±	0.291	<0.0017		<0.0025	0.004 ±	0.001
Sam	0.0019 ±	0.0002	0.013 ±	0.001	9.403 ±	0.068	1.518 ±	0.520	<0.0017		<0.0025	0.009 ±	0.001
Hayawiyia	0.0027 ±	0.0010	0.084 ±	0.011	59.169 ±	0.174	11.847 ±	0.854	0.0536 ±	0.0036	<0.0025	0.265 ±	0.021
Lojina	<0.0005		0.010 ±	0.001	0.346 ±	0.031	0.793 ±	0.130	<0.0017		<0.0025	0.016 ±	0.001
Haddain	<0.0005		0.013 ±	0.002	0.372 ±	0.022	7.252 ±	0.662	<0.0017		<0.0025	0.008 ±	0.001

Alalam	0.0034 ±	0.0003	0.207 ±	0.019	0.259 ±	0.037	1.155 ±	0.077	<0.0017		<0.0025	0.006 ±	0.001
Alqemmah	0.0049 ±	0.0004	< 0.003		0.702 ±	0.012	1.340 ±	0.142	<0.0017		<0.0025	0.024 ±	0.002
Watani	0.0018 ±	0.0001	0.028 ±	0.004	<0.036		2.221 ±	0.113	<0.0017		<0.0025	0.016 ±	0.001
Abar	0.0122 ±	0.0010	0.018 ±	0.001	0.470 ±	0.040	3.149 ±	0.091	0.0032 ±	0.0005	<0.0025	0.006 ±	0.001
Riwa	<0.0005		< 0.003		<0.036		3.262 ±	0.202	<0.0017		<0.0025	0.006 ±	0.001

3.2.2. Aluminum

In toxicological investigations, aluminum (Al) has been found to be neurotoxic [27], [28]. Moreover, there are conflicting studies regarding the relationship between aluminum in drinking water and Alzheimer's disease. Some studies support the connection, while others refute it [29], [30], [31]. The Alhodhod sample had the highest measured concentration of Al, with a value of 5.810μg/L, while the Hana sample had the lowest content at 0.081μg/L. Comparing these values to the MAC set for Al, it is clear that the aluminum content in all tested brands is significantly lower than the limits specified by Yemeni regulations, as shown in Table 4.

3.2.3. Vanadium

The concentration of vanadium (V) in drinking water can reach levels as high as 100μg/L [32]. However, the average concentrations typically fall within the range of 1 to 6μg/L [33]. In the examined bottled drinking water samples of this study, the concentrations of V ranged from 0.051 to 18.803μg/L.

3.2.4. Chromium

The risks associated with chromium (Cr) to the environment and human health are already well-known under specific conditions [34]. As a result, both the WHO and the laws in Yemen have established a MAC value for Cr in drinking water. This MAC value is consistent in both documents and is set at 50μg/L [21], [22]. Cr was found in all the examined samples, with the highest concentration of 1.263μg/L observed in the Mozn sample.

3.2.5. Manganese

Excessive levels of manganese (Mn) in drinking water, particularly those above 100μg/L, can lead to various issues such as unpleasant taste, staining of sanitary ware and laundry, and the formation of a black coating on pipes [35]. Furthermore, there is increasing evidence suggesting that Mn concentrations exceeding 100μg/L in drinking water pose a notable neurotoxic risk, especially for children. [36]. Consequently, the WHO has not set a MAC value for it [21]. YMWE sets the MAC of Mn in drinking water at 200μg/L. [22]. In all the tested bottled drinking water samples, the presence of Mn was found to be below the limit of detection, which is<0.036μg/L.

3.2.6. Iron

Iron (Fe) in drinking water at normal levels is not harmful to human health. WHO does not provide a specific guideline for the quantity of Fe in drinking water due to its low health risks, the YMWE has set a MAC value for iron in drinking water. The MAC value is established at 100μg/L. From the data presented in Table 4, it can be observed that nearly half of the analyzed samples have Fe levels below the limit of detection. The remaining samples exhibit Fe concentrations ranging from 0.087 to 33.300μg/L.

3.2.7. Cobalt

Cobalt (Co) is characterized by a low level of toxicity. Moreover, the concentration of cobalt in drinking water is generally low, typically falling within the range of 0.1 to 5μg/L [37]. Therefore, no specific MAC values have been set for Co in drinking water [21], [22]. Table 4 illustrates that the cobalt levels in the analyzed samples ranged from a minimum of 0.009μg/L in the Hana sample to a maximum of 0.218μg/L in the Alkhayr sample. The average concentration of cobalt in the studied samples is 0.07μg/L.

3.2.8. Nickel

Nickel (Ni) is considered one of the toxic elements that can potentially increase the risk of cancer in humans [38]. The following rules govern the MAC of Ni in drinking water: 70μg/L by the WHO [21], compared to 20.0μg/L by YMWE [22]. In some of the bottled water samples, nickel was not found. In the samples where it was present, its concentration was generally below 0.386μg/L. This concentration is around 51.8 times lower than the drinking water standards set by the YMWE.

3.2.9. Copper

Copper (Cu) plays a dual role in drinking water as both an essential nutrient and a potential pollutant. Elevated Cu levels in drinking water are frequently caused by factors like high mineral content in groundwater or the use of corrosive pipes and plumbing materials [21]. The WHO and YMWE have established MAC values for copper in drinking water at 2000μg/L and 1000μg/L, respectively. Analysis of bottled water samples showed copper concentrations below 1.312μg/L.

3.2.10. Zinc

Zinc (Zn) is a necessary trace element for the health of plants, animals, and humans. However, high levels of Zn can lead to contamination of soil, water, and the food chain. Drinking water that exceeds Zn concentrations of 3000μg/L may have an unpleasant taste and appearance [39]. The WHO does not specify a MAC value for zinc in drinking water. However, the YMWE advises that zinc concentrations should not exceed 15000μg/L. In the evaluation of Yemeni bottled waters, it was found that Zn was present in most samples below the specified LOD (1.860μg/L). Zn was detected in three specific samples, namely Mozn, Ghailan, and Abar, with concentrations of 7.834μg/L, 6.968μg/L, and 3.213μg/L, respectively.

3.2.11. Gallium

Gallium (Ga) is classified as a non-essential metallic trace element in terms of its role in supporting biological life processes [40], [41]. The disposal of waste containing gallium by electronic industries can pose substantial health risks to humans. Therefore, it is crucial to monitor Ga levels in order to assess water quality [42].

However, neither the WHO nor Yemeni legislation has provided specific MAC values for Ga in drinking water. Table 4 shows varying Ga concentrations in the water samples. The lowest recorded level was 0.003 µg/L in the Hana, Nofan, Haddain, and Riwa samples, while the Hayawiyia sample had the highest concentration at 0.851 µg/L.

3.2.12. Selenium

Selenium (Se) is a vital element for human health. A deficiency in Se can result in heart muscle failure and a condition called white muscle disease [21]. On the other hand, high oral exposure to Se can lead to acute toxicity [43]. The WHO and the YMWE have established different MAC values for Se in drinking water. The WHO recommends a MAC value of 40 µg/L, while the YMWE sets a stricter MAC value of 10 µg/L. The levels of Se in the evaluated bottled water samples for this study ranged from 1.500 to 4.828 µg/L, with an average concentration of 3.339 µg/L.

3.2.13. Rubidium

Rubidium (Rb), an alkali metal, has been studied as an essential element for the human body [44], [45]. The WHO and Yemeni standards do not specify MAC values for Rb in drinking water. In the tested bottled water samples, Rb concentrations ranged from 0.024 to 1.647 µg/L, with an average concentration of 0.664 µg/L.

3.2.14. Silver

Silver (Ag) concentrations above 5 µg/L have been detected in groundwater, surface water, and drinking water. Recent estimates suggest that individuals may consume around 7 µg of silver per day. [21]. There is no MAC value for Ag in drinking water according to the WHO or Yemeni standards. In Table 4, Ag concentrations in some samples were below the detection limit (0.005 µg/L), while in other samples, Ag was detected within the range of 0.012–0.076 µg/L.

3.2.15. Cadmium

Cadmium (Cd) is a hazardous element that can be found in occupational settings as well as the environment. Following absorption, Cd accumulates within the human body, primarily in the liver. This accumulation can lead to severe liver damage, including conditions such as steatosis (fatty liver), steatohepatitis (inflammation of the liver with fat accumulation), and, ultimately, hepatocellular cancer. [46]. The WHO and YMWE have established MAC values for Cd in drinking water: 3 µg/L and 5 µg/L, respectively [21], [22]. The Cd concentration in most analyzed samples ranged from 0.0013 to 0.1421 µg/L. The Sana'a sample had the highest concentration, while the Hana, Hayat and Alkhayr samples had the lowest. Furthermore, four bottled drinking water samples had Cd concentrations below the detection limits, as shown in Table 4.

3.2.16. Cesium

The toxicity of cesium (Cs) to humans is rarely mentioned [47], and as a result, the WHO and YMWE have not prescribed any MAC value for Cs in drinking water [21], [22]. The Alalam sample exhibited the highest concentration of Cs in the water samples, reaching 0.207 µg/L, while several other samples remained below the detection limits, as indicated in Table 4.

3.2.17. Barium

Barium (Ba) is an alkaline earth metal, and cases of poisoning related to it are uncommon [48]. The WHO recommends that the concentration of Ba in drinking water should not exceed 1300µg/L, while the Yemeni legislation does not specify a MAC value for Ba. The concentrations of Ba in all the analyzed bottled water samples for this study varied between 0.152 and 59.169µg/L, except for the Watani sample, where the concentration of Ba was below the detection limits, as indicated in Table 4.

3.2.18. Mercury

Mercury (Hg) is a common, extremely toxic, persistent contaminant with harmful effects on humans [49]. Upon exposure, mercury ions accumulate in the brain and kidneys, leading to detrimental effects on various organ systems, including the renal, nervous, cardiovascular, and musculoskeletal systems [50], [51], [52]. The WHO and the YMWE recommend that the concentration of Hg in drinking water should not exceed 6µg/L and 1µg/L, respectively. The concentrations of Hg in all the samples analyzed ranged from 0.649 to 15.492µg/L. The Sana'a sample has the highest concentration of 15.492µg/L, while the Alkhayr sample has the lowest concentration of 0.649µg/L. The concentration of Hg in all the examined samples exceeded the guideline value specified in the Yemeni standards, with the exception of the Alkhayr and Lojina samples. Furthermore, a quarter of the samples surpassed the guideline value set by the WHO, as shown in Table 4. The bottled water samples that exceed the guideline value for Hg concentration set by the WHO include Sana'a, Hana, Hayawiyia, Yemen, Ghuom, Haddain, and Hadda samples. A comparison of mercury levels in all the samples is depicted in Fig. 1.

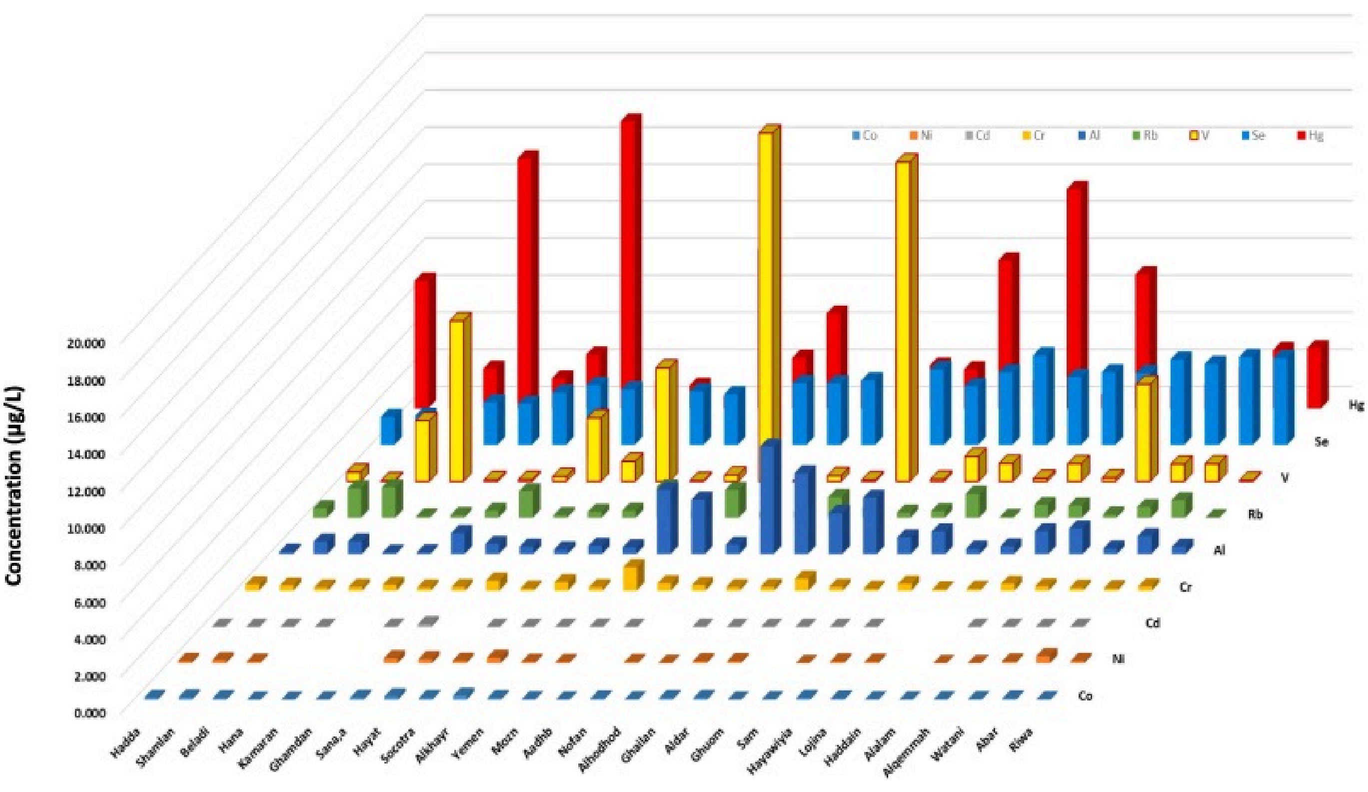

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Fig. 1. Distribution of Co, Ni, Cd, Cr, Al, Rb, V, Se, and Hg in 27 bottled drinking waters on the Yemeni market.

3.2.19. Thallium

Thallium (Tl) is a highly toxic heavy metal. Its toxicity to humans is comparable to that of Hg, Cd, or Pb [53], [54]. The WHO and Yemeni standards do not specify MAC values for Tl in drinking water. However, according to the U.S. Environmental Protection Agency (EPA), the guideline value for Tl in drinking water is 2µg/L.[23]. Prolonged exposure to Tl concentrations that surpass the guideline value of 2µg/L, as set by the U.S. EPA, can lead to adverse effects, including hair loss, blood abnormalities, and complications affecting the kidneys, intestines, or liver [23]. The concentration of Tl in the majority of the analyzed bottled drinking water samples was found to be below the detection limits, as shown in Table 4. However, eleven bottled water samples showed concentrations ranging from 0.0023 to 0.0536µg/L. It is important to note that the detected Tl content in the studied samples is significantly below the MAC, according to the U.S. EPA.

3.2.20. Lead

Lead (Pb) poisoning and its cumulative effects have profound health implications for various bodily systems, including the brain, kidneys, blood, central nervous system, and reproductive system [55], [56], [57]. As per the guidelines provided by the WHO and YMWE, the MAC levels of Pb in drinking water are 10µg/L and 5µg/L, respectively [21], [22]. Upon examination, all analyzed samples of Yemeni bottled water exhibited Pb concentrations below the detection limit of 0.0025µg/L. This signifies that the Pb levels in the tested samples are considerably lower than the permissible limit for Pb in drinking water.

3.2.21. Uranium

Uranium (U) is not considered an essential element for any organism and has not been shown to have any biological function in the human body [58], [59]. On average, humans ingest approximately 2.6µg of uranium per day, with drinking water contributing around one-third of this intake, based on the average weight of 70kg for an adult male. Uranium primarily affects the kidneys, and prolonged exposure to this element can lead to neurological behavior changes [60]. The WHO has established a MAC of 10µg/L for U in drinking water. However, Yemeni regulations do not specifically address this MAC value. In the analyzed samples of bottled water, U was detected at concentrations ranging from 0.002 to 0.265µg/L, with an average concentration of 0.038µg/L. These concentrations were considerably lower than the MAC level set by the WHO [21].

4. Conclusion

Using ICP-MS, 21 trace and heavy metals were analyzed in 27 samples of bottled drinking water from Yemen's market. The analysis results of the studied samples indicated that the concentrations of the majority of elements were well below the MAC as specified by the WHO and YMWE. In addition, the TDS, EC, and pH levels were within the standards set by the YMWE and the US EPA. Notably, Mn and Pb were undetectable in all samples, while Zn, Ag, and Tl were mostly below the detection limits. On the other hand, Hg is the only element that demonstrated levels surpassing Yemeni standards in most samples and exceeding WHO guidelines in some samples. The concentrations of Hg in the Sana'a, Hana, and Hayawiyia samples were 15.492, 13.506, and 11.847µg/L, respectively. These values are approximately double or higher than the MAC specified in the WHO guidelines and more than eleven times higher than the limits set by the Yemeni guidelines. To ensure the provision of safe drinking water for the people in Yemen, it is crucial to conduct research to identify the source of mercury contamination in bottled water.

CRediT authorship contribution statement

Ibrahim A. Alhagri: Writing – review & editing, Formal analysis, Data curation. **Ahmed N. Al-Hakimi:** Writing – original draft, Validation, Investigation. **Sadeq M. Al-Hazmy:** Writing – original draft, Resources, Investigation. **Abuzar E.A.E. Albadri:** Visualization, Project administration, Methodology, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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