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Chemical, microbial and physical evaluation of commercial bottled waters in greater Houston area of Texas

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Due to the increased demand and consumption of bottled water in the United States, there has been a growing concern about the quality of this product. Retail outlets sell local as well as imported bottled water to consumers. Three bottles for each of 35 different brands of bottled water were randomly collected from local grocery stores in the greater Houston area. Out of the 35 different brands, 16 were designated as spring water, 11 were purified and/or fortified tap water, 5 were carbonated water and 3 were distilled water. Chemical, microbial and physical properties of all samples were evaluated including pH, conductivity, bacteria counts, anion concentration, trace metal concentration, heavy metal and volatile organics concentration were determined in all samples. Inductively coupled plasma/mass spectrometry (ICPMS) was used for elemental analysis, gas chromatography with electron capture detector (GCECD) as well as gas chromatography mass spectrometry (GCMS) were used for analysis of volatile organics, ion chromatography (IC) and selective ion electrodes were used for the analysis of anions. Bacterial identification was performed using the Biolog software (Biolog, Inc., Hayward, Ca, USA). The results obtained were compared with guidelines of drinking water recommended by the International Bottled Water Association (IBWA), United States Food and Drug Administration (FDA), United States Environmental Protection Agency (EPA) and the World Health Organization (WHO) drinking water standard. The majority of the analyzed chemicals were below their respective drinking water standards for maximum admissible concentrations (MAC). Volatile organic chemicals were found to be below detection limits. Four of the 35 brands of the bottled water samples analyzed were found to be contaminated with bacteria.

Keywords: Bottled water, water quality constituents, inductively coupled plasma (ICPMS), drinking water standards, maximum admissible concentrations (MAC).

Introduction

According to the International Bottled Water Association (IBWA),[1] drinking water has become extremely popular with a current U.S. market of more than 11 billion American dollars. Its consumption has tripled in the past 10 years, making it the second largest beverage product category, behind soft drinks. USA consumption in the year 2006 was 32 billion liters compared to 20 billion liters in the year 2001. The average number of liters consumed by person per year in the USA is 90.5 and the global average is 24.2 liters. Such explosive growth in the bottled water industry is presumably a result of people's perception of purity, safety, better taste, convenience, and increasing public awareness of fitness and beneficial effects of drinking water on health. More than one-half of all Americans drink bottled water, and approximately one-third of the public consumes it regularly.[2] Bottled water is regulated by the International Bottled Water Association (Alexandria, Virginia). However, regulations are less stringent compared to U.S. Environmental Protection Agency (U.S. EPA) regulations for tap water.[3,4] Bottled water is considered a food product and is also regulated by the Food and Drug Administration (FDA) (Rockville, Maryland). All bottled water products must comply with FDA's quality standards, labeling regulations, and Good Manufacturing Practices.[1,4]

Recently, an increasingly worldwide concern about the quality of bottled water regarding their chemical contents has risen.[5–9] Water quality can be measured by means of its concentration in organic and inorganic chemicals. Mineral content of bottled water is one of the most important markers for water quality. Some minerals are required by our bodies for numerous biological and physiological processes that are necessary for the maintenance of health. These elements are divided into two categories: those that are required in our diet in amounts greater than 50 mg per

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day are designated as macro elements, and those that are required in amounts less than 50 mg per day, which are called trace elements. Despite the fact that trace elements constitute only a small fraction of the total food uptake, epidemiological studies have shown a strong correlation between several human diseases and the presence of trace elements in drinking water. Also there is an increasing concern about the microbial quality of bottled water marketed worldwide. Several studies have documented the detection of coliforms and heterotrophic bacteria in bottled water in counts which far exceeded national and international standards set for potable water for human consumption.[10–12] Also of importance are reports of potential pathogens, such as enteric bacteria,[13] protozoa,[14–15] and acid-fast bacteria.[16,17] It has also been established that a number of these bacteria could multiply during storage to reach infective doses for consumers.[18,19] Pathogens like *Escherichia coli, Pseudomonas* spp and *Salmonella* spp have been demonstrated to survive and multiply in bottled water with a potential to cause outbreaks in consumers.[20,21] As a part of our ongoing research on the assessment of human exposure to toxic chemicals, chemical, physical and microbial evaluations of local bottled drinking water were conducted. This paper discusses the levels of trace elements and other toxic pollutants in 35 different brands of imported and domestic bottled water sold in the greater Houston area of Texas.

Material and methods

Sample collection

One hundred and five bottles representing thirty five brands of bottled water were purchased randomly from different supermarket stores in Houston, Texas, USA during the summer of 2005. All samples were in plastic containers with plastic screw caps. Table 1 presents the classification of the bottled waters in terms of brands, types, sources and volumes.

Reagents

All chemicals used were of reagent grade and ultra pure blank water was purchased from Fisher Scientific (Suwannee, GA, USA). One hundred mg/L of mixed standard solutions of anions and cations were obtained from SPEX Certiprep, Inc (Metuchen, NJ, USA). Metal standards used were purchased from Fisher Scientific. The ICPMS was calibrated by preparing calibration standard solutions from the stock standard solutions and an internal standard was used as part of the quality assurance. Standard reference materials (SRM 1640: trace elements in natural water) was purchased from National Institute of Standards and Technology (NIST), Gaithersburg, MD, USA.

Analytical methodology and quality assurance

Water quality constituents were analyzed following standard analytical methods for the examination of water and wastewater.[22] Analytical techniques and detection limits of analytes are shown in Table 2.

pH, conductivity, and ion selective electrodes (ISE) analysis

Water samples used for ion selective electrodes analysis were mixed with the ionic strength adjustor of the selected ISE. The ion selective electrodes used were cyanide combination electrode (Model 96-06), ammonia electrode (Model 95-12), combination fluoride electrode (Model 96-09), and nitrite electrode (Model 9346) with a single junction reference electrode model 90-01 (all electrodes were from Orion, MA). A conductivity cell (Model 0 17010) and a pH meter were also used. These electrodes were connected to an EA907 expandable ion analyzer (Orion, MA). Ion selective electrodes were checked, filled with their filling solution and measured for electrode slope, where the slope was defined as the change in millivolts per 10-fold change in concentration. The difference between the two readings was the slope of the electrode. Calibrating standards for ISE were made up from 1000 ppm ready-made stock solutions purchased as certified reference standards from Fisher Scientific Inc. NH, USA.

Inductively coupled plasma mass spectrometry (ICPMS)

All samples were acidified with ultra pure nitric acid to pH 2. One hundred milliliter aliquots of the acid preserved samples were analyzed by ICPMS using a Hewlett-Packard (HP) 7500C ICPMS equipped auto sampler and shield torch octapole reaction system (Hewlett-Packard Co., Wilmington, DE) according to the EPA Method 200.8.[23] Quality assurance and control of data were performed according to the specified condition of the method and consisted of analysis of laboratory reagent blanks, fortified blanks and samples as a continuing check on performance. Rinse blanks and three standard solutions of all monitored elements were used at concentrations of 10, 100 and 400 μg/L (ppb). Two calibrated standard solutions at concentration of 1 and 5 ppb were used for mercury determinations.

Ion chromatography

Ion chromatography was carried out using a Dionex gradient HPLC system (Sunnyvale, CA) equipped with a CD20 conductivity detector. Isocratic separations of both cations and anions were performed on CS12 and AS 14 analytical columns, respectively. Dedicated guard columns and suppressor systems to either cationic or anionic analyses were also used in connection with the analytical columns. Methanesulphonic acid (20 mM) was used as the mobile phase for eluting cations, while a mixture of 3.5 mM

Table 1. Bottled water classification, sources and volume

Sample #	*Brand Names*	*Source*	*Volume*
I. Spring Water			
	Texas Water		
1	Natural Spring Water, HEB, San Antonio, TX 78204	Samantha Springs, Keller, Texas	237 mL
2	Refresh Natural Spring Water, Safeway Inc. PO Box 99, Pleasanton, California, 94566-0009	Samantha Springs, Keller, Texas	500 mL
3	Nestle Natural Spring Water, Nestle Waters Canada, Guelph, Ontario, Canada NIH 6H9	Clear Spring, Wood County, Texas	500 mL
4	Ozarka Natural Spring Water, Nestle Waters, North America Inc., Greenwich, CT 06830	Piney Woods Springs, Wood County, TX	237 mL
5	Remarkable Spring Water, Randall's/Tom Thumb Food Markets, Houston, TX 77210	Buck Springs Inc. Jasper, TX 75951	3.78 L
	Pennsylvania Water		
6	Pure American Natural Spring Water, CCDA Waters, L.L.C. Atlanta, GA 30313	Big Spring, Bellefonte, PA	240 mL
7	Dannon Fluoridated Spring Water, CCDA Waters, L.L.C. Atlanta, GA 30313	Big Spring, Bellefonte, PA	255 mL
	Florida Water		
8	Walgreen's Spring Water, CCDA Waters, L.L.C. Atlanta, GA 30313	Ginnie Spring, Gilchrist CTY, High Springs, FL	500 mL
9	Dannon Natural Spring Water, CCDA Waters, L.L.C. Atlanta, GA 30313	Ginnie Spring, Gilchrist CTY, High Springs, FL	1 L
	Louisiana Water		
10	Kroger Spring Water, Kroger Co, Cincinnati, OH 45202	Natural Spring in Webster Parish, LA	3.78 L
11	Fiesta Spring Water, 5235 Katy Fwy, Houston, TX 77007	Natural Spring in Webster Parish, LA	3.97 L
	Arkansas Water		
12	Music Mountain Arkansas Spring Water Co., 1078 Big Fork Road, Norman, AR 71953	Wilderness Valley Springs	3.97 L
	Tennessee Water		
13	Crystal Geyser Natural Alpine Spring Water, Mountains of Tennessee, 37307	Alpine Spring Water, Benton, TN	500 mL
	Ohio Water		
14	Kroger Spring Water, Kroger Co, Cincinnati, OH 45202	Ouachita National Forest, Montgomery, AR	500 mL
	Imported Water		
15	Glacia Natural Canadian Spring Water, HEB, San Antonio, TX 78204	Osprey TWP, Ontario, Canada	500 mL
16	Evian Natural Spring Water, S.A. Evian CO, At Avian 74503 France	Cachat Spring, Evian, France, French Alps	330 mL
II. Municipal Water			
	Purified Municipal Water Fortified With Minerals		
17	Sparkletts Crystal-Fresh Purified Water, Micron filtered, reverse osmosis, ozonated, and fortified with sodium bicarbonate, sodium sulfate, magnesium chloride and calcium chloride	CCDA waters, LLC, Atlanta, GA 30313	500 mL
18	Dasani Purified Water, Reverse osmosis purified and filtered. Enhanced with minerals	Coca Cola Company	1 L
19	Arctic Mist Purified Water, Reverse osmosis purified and filtered. Enhanced with minerals	CCDA Waters, L.L.C. Atlanta, GA 30313	1.5 L
20	Ozarka Drinking Water, Micron filtered, fortified with calcium chloride, magnesium chloride and sodium bicarbonate	Nestle Waters North America Inc., Greenwich, CT 06830	3.97 L

(continued on next page)

Table 1. (*Continued*)

Sample #	*Brand Names*	*Source*	*Volume*
	Purified Municipal Water, Houston, Texas		
21	Clear Mist Purified Drinking Water, Filtered and ozonated	CCDA Waters, L.L.C. Atlanta, GA 30313	550 mL
22	Deja Blue Purified Drinking Water, Reverse osmosis purified, carbon filtration and ozonated	CCDA Waters, L.L.C. Atlanta, GA 30313	1 L
23	Aquafina Purified Drinking Water, non-carbonated purified drinking water, reverse osmosis and ozonated	CCDA Waters, L.L.C. Atlanta, GA 30313	1 L
24	Fiesta Drinking Water, Deep Well in Shreveport, LA, Drinking water, carbon filtered, reverse osmosis, micron filtered and ozonated	Fiesta Drinking Water	3.97 L
25	Oasis Drinking Water, Purified through carbon filtration, reverse osmosis and ozonated	Nestle Waters North America Inc., Greenwich, CT 06830	3.97 L
26	Kroger Drinking Water, Drinking water carbon filtration, microfiltration and ozonated	Kroger Co, Cincinnati, Ohio 45202	3.97 L
27	Remarkable Drinking Water, Sodium free drinking water microfiltration and ozonation	Randall's/Tom Thumb Food Markets, Houston, TX 77210	3.7 L
III. Carbonated Water			
28	Crystal Clear Sparkling Water Inter-American Products, Inc.	Cincinnati, Ohio 45202	1 L
29	Big K Sparkling Water American Products, Inc. Cincinnati, Ohio 45202	Cincinnati, Ohio 45202	2 L
30	La Croix Pure Sparking Water Ever fresh/lacroix beverages Inc., Warren, MI 48091	Warren, MI 48091	1 L
31	Crystal Geyser Sparkling Mineral Water Crystal Geyser Water Co., Benton, TN	San Francisco, CA 94133	1L
32	Perrier Sparkling Natural Mineral Water Perrier Sparkling, Vergeze, France	Mineral Water, Vergeze, France	3.97 L
33	Vapor distilled and fortified with electrolytes, calcium, magnesium chlorides and potassium bicarbonate	Glaceau Smart Water Energy brands Inc, Whitestone, NY 11357	1.5 l
34	Distilled water carbon filtration, microfiltration and ozonated	Kroger Distilled Water Kroger Co., Cincinnati, Ohio 45202	3.97 L
35	Purified by steam distillation, sodium-free carbon filtration, microfiltration and ozonation	Ozarka Distilled Water Nestle Waters North America Inc., Greenwich, CT 06830	3.97 L

Na_2CO_3/1.0 mM $NaHCO_3$ was used as the mobile phase for eluting the anions. Data acquisition and instrument settings were performed by Peaknet software (Dionex, CA). Primary calibration standard solutions (1000 ppm) for ions were prepared from their ultra-pure salts. Working standard solutions were prepared from the primary solutions following proper serial dilutions. The concentrations were measured using external calibration method and the analyses were carried out in triplicate and the average values were reported. Instrument precisions was determined by introducing the same quantity of one sample 10 times, then the relative standard deviation was calculated and found to be less than 8%.

Gas chromatography (GC) and gas chromatography/mass spectrometry (GC/MS) analysis

Trihalomethanes were determined using headspace GC and electron capture detection according to the USEPA method 8010 using an HP 5890 Series II plus gas chromatograph attached to an HP 7694 headspace sampler and a 63Ni electron capture detector. Conformation of the GC results was carried out using an HP 5972 mass selective GC/MS system.

Microbiological analysis

Microbial contamination in the tested bottled waters were examined by filtration of samples through nitrocellulose membranes (0.45 μm pore size, 47 mm diameter, Pall-Gelman Laboratory) followed by plating on selective media according to Reasoner.[24] A 250 mL of each sample was filtered through the membrane filter plated on agar followed by incubation at 37 °C for 48 h. All colonies were counted as colony forming units per milliliter of the water sample. The isolated microorganisms were identified according to their biochemical characteristics and using standardized identification systems of BIOLOG Inc (Howard, California).

Table 2. Analytical techniques that were used for water analysis and their detection limits (μg/L)*

	Analytical Techniques		
Analytes	*Inductively Coupled Plasma/Mass Spectrometry (ICPMS)*	*Ion Selective Electrode (ISE)*	*Ion Chromatography (IC)*
Na	5.0	20	200
K	6.0	40	200
Ca	6.0	20	100
Mg	5.0		200
Al	0.3		
NH_4		10	7.0
Cl		1800	170
SO_4			100
NO_2		20	100
NO_3		100	60
F		20	190
PO_4			250
CN		200	50
Be	0.02		
Ti	0.01		
V	0.01		
Cr	0.01		
Mn	0.15		
Fe	10.0		
Co	0.01		
Ni	0.01		
Cu	0.01		
Zn	0.01		
As	1.0		
Se	0.06		
Mo	0.01		
Ag	0.05		
Cd	0.01	10	
Sn	0.04		
Sb	0.005		
Ba	1.0		
Hg	0.02		
Tl	0.01		
Pb	0.001	200	
B	1.0		
Si	1.0		
Sr	0.001		

*Detection limit for halomethans was 3 μg/l using head space GC/ECD technique.

Results and discussion

The concentrations of major cations, anions, conductivity, pH and bacterial counts in bottled water samples are shown in Table 3. The numbers shown are the average measurements of three replicates for each of water brand. Standard deviation among replicates was found to be less than 10% of the mean values. Drinking water recommended standards set by the IBWA, FDA, EPA and the WHO are shown in Table 4.

Physical properties

The pH of the majority of the analyzed bottled waters were in the range of ranged from 7 to 8 with the exception of carbonated waters which were much more acidic with a pH ranging from 4.35 to 5.60. recommended pH values for drinking water according to (Table 4) are 6.5 to 8.5; only carbonated waters, 3 brands of spring water and one brand of distilled water were out of that recommended range. At pH values less than 6.5, water is corrosive and dissolves plumbing components. High pH values (8.5 or more) can promote hardness scale precipitation and make chlorine disinfectants more effective. Electric conductivity (EC) generally reflects total dissolved solids in drinking water. It was found to be the highest for carbonated waters, followed by spring waters, tap waters and was lowest for distilled waters. Only carbonated water brands and 2 of the spring water brands exceeded the EPA/WHO recommended values. An elevated TDS concentration is not a health hazard. It

Table 3. Major cations concentration, anions concentration and physical properties of bottled waters

I. Spring Water

	Cations mg/L (ppm)					Anions mg/L (ppm)				Physical and Biological		
Sample #	Na^+	K^+	Mg^{2+}	Ca^{2+}	F^-	Cl^-	NO_2-	NO_3-	SO_4^{2-}	*Conductivity*	*pH*	*Bacteria*
Texas Water												
1	34.60	3.20	15.45	116.58	0.1	29.7	BDL	26.22	40.7	375	7.89	None
2	29.27	3.24	16.10	134.74	0.1	0.1	BDL	22.45	47.1	421	7.77	>50[a]
3	8.34	4.23	4.74	17.80	BDL	4.3	BDL	7.51	3.9	94	5.93	None
4	5.85	0.64	1.41	7.61	2.4	BDL	10.1	BDL	BDL	55	7.37	None
5	5.31	0.79	0.73	1.81	0.1	2.6	BDL	50.01	0.6	39	6.26	None
Pennsylvania Water												
6	14.08	0.52	34.06	114.57	0.3	15.6	0.3	8.23	9.6	349	7.96	>30[a]
7	15.81	1.51	34.39	115.99	0.7	15.6	0.2	8.42	8.8	362	7.92	21[b]
Florida Water												
8	4.58	0.20	8.26	152.59	0.1	3.6	BDL	5.44	7.5	345	6.67	None
9	4.73	0.39	8.46	156.01	0.1	3.4	BDL	4.51	7.4	345	6.38	12[c]
Louisiana Water												
10	10.91	6.47	3.88	17.82	BDL	4.0	BDL	33.91	6.9	103	6.68	None
11	10.24	6.44	3.78	8.65	BDL	4.5	BDL	16.02	8.2	168	7.37	50[a,b]
Arkansas Water												
12	5.47	0.85	3.74	173.55	0.1	6.4	BDL	35.04	7.5	331	7.26	None
Tennessee Water												
13	5.56	1.16	9.92	124.94	0.1	3.5	BDL	5.61	4.6	305	7.10	None
Ohio Water												
14	5.18	BDL	3.51	172.70	0.2	2.7	BDL	2.35	5.2	361	7.55	None
Imported Water												
15	3.95	0.77	56.30	245.83	0.1	2.7	BDL	7.81	6.2	586	7.71	None
16	11.83	BDL	54.78	225.89	BDL	5.4	BDL	3.53	13.3	502	7.78	5[f]

[a] *Klebsiella terrigena* (Gram Negative), [b] *Ralstonia pickettii* and *Acidovorax temperans* (Gram Negative), [c] *Acidovorax delafieldii* (Gram Negative), [d] *Burkholderia glumae* (Gram Positive), [e] *Bacillus thermoglucosidasius* (Gram Positive), [f] *Agrobacterium rhizogenes* (Gram Negative), [g] *Moraxella caviae* (Gram Negative).

II. Municipal Water

Sample #	Na^+	K^+	Mg^{2+}	Ca^{2+}	F^-	Cl^-	NO_2^-	NO_3^-	SO_4^{2-}	CN^-	*Conductivity*	*pH*	*Bacteria*
Purified Municipal Water Fortified With Minerals													
17	7.15	0.07	0.02	0.14	0.1	0.9	BDL	0.41	5.3	0.2	59	8.0	40[f,g]
18	4.21	3.94	4.18	9.84	BDL	6.5	BDL	0.24	11.3	0.6	77	6.67	None
19	4.13	BDL	0.63	17.38	BDL	0.7	BDL	0.18	BDL	BDL	58	7.21	None
20	9.10	2.21	7.67	21.38	0.1	BDL	BDL	1.11	1.9	BDL	184	7.81	None
Purified Municipal Water Houston, Texas													
21	1.64	0.03	BDL	0.31	BDL	0.9	0.03	0.09	0.4	0.1	30	6.94	50[f,g]
22	2.87	0.26	0.50	1.88	BDL	0.9	BDL	0.23	BDL	BDL	38	7.67	None
23	3.14	BDL	0.17	1.01	BDL	1.6	BDL	0.34	0.4	BDL	75	7.35	None
24	5.01	0.21	0.08	1.17	BDL	1.2	BDL	0.14	BDL	BDL	21	8.13	None
25	6.08	0.87	0.52	7.72	0.1	3.9	BDL	0.43	3.1	BDL	84	7.43	None
26	5.25	0.20	0.07	0.92	0.7	2.6	BDL	0.25	BDL	BDL	38	7.29	30[f,g]
27	5.30	0.83	0.78	0.82	BDL	4.4	0.01	0.94	0.7	BDL	98	7.06	None

Only sample # 23 has 0.059 ppm ammonia NH_4^+.

(Continued on next page)

Table 3. *(Continued)*

III. Carbonated Water

Sample #	Na^+	K^+	Mg^{2+}	Ca^{2+}	F^-	Cl^-	NO_2^-	NO_3^-	SO_4^{2-}	CN^-	PO_4^{3-}	*Conductivity*	*pH*
28	39.66	7.22	4.90	171.55	0.6	30.4	0.52	10.16	161.1	0.1	0.2	523	4.35
29	38.57	7.08	4.74	152.32	0.4	25.9	0.30	13.33	141.1	BDL	0.1	490	4.38
30	12.91	2.94	2.23	48.30	0.6	5.5	BDL	12.99	9.0	BDL	0.1	178	4.54
31	175.97	14.37	37.81	15.75	0.6	40.8	BDL	4.57	3.9	0.1	BDL	842	5.27
32	19.66	1.66	13.25	514.60	0.1	22.3	BDL	15.05	56.4	BDL	BDL	878	5.60

Only Sample 28 has 0.328 ppm NH_4^+; only sample 31 has 0.112 ppm Br^-; No bacteria were found in any samples.

IV. Distilled Water

Sample #	*Li*	Na^+	*NH4*$^+$	K^+	Mg^{2+}	Ca^{2+}	F^-	Cl^-	NO_2^-	NO_3^-	SO_4^{2-}	CN^-	*Conductivity*	*pH*
33	BDL	7.51	BDL	7.44	5.99	16.11	BDL	22.6	BDL	0.29	0.3	BDL	118	6.83
34	BDL	2.35	BDL	0.52	0.36	2.84	BDL	1.3	0.01	0.17	0.5	BDL	21	5.59
35	0.05	2.38	0.10	0.60	BDL	BDL	BDL	1.1	0.01	0.39	0.2	BDL	54	9.00

was regulated by WHO guidelines because it is more of an aesthetic rather than being harmful to health.

Microbial investigation

Six brands of spring waters showed minor contamination with bacteria with counts in the range of 5–50 bacteria/mL, 3 brands of bottled tap water with a range of 30–50 bacteria/mL, none of the carbonated water or distilled water brands were contaminated with bacteria. Identification of the bacteria was carried out using the automated Biolog program as described in the experimental section. Identified bacteria in spring water brands were 4 Gram-negatives, *Klebsiella terrigena, Ralstonia pickettii,* C*idovorax temperans,* and *Acidovorax delafieldii* and *Agrobacterium rhizogenes*, 2 Gram-positives *Burkholderia glumae* and *Bacillus thermoglucosidasius.* Purified tap waters showed only 2 types of Gram-negative bacteria, *Burkholderia glumae* and *Moraxella caviae.*

Major cations

Sodium, potassium, magnesium and calcium concentrations in the bottled water samples were analyzed by ion chromatography as shown in the experimental section. The results are shown in Table 3. Sodium (Na) concentrations varied between 12.9 to 175.9, 3.9 to 34.6, 1.6 to 9.1, and 2.3 to 7.5 mg/L for carbonated waters, spring waters, tap waters and distilled water, respectively. None of the values exceeded the sodium maximum limit of 200 mg/L (Table 4) guidelines for aesthetic quality. Most water contains some sodium, which naturally leaches from rocks and soils. An excess of sodium more than 200 mg/L in drinking water may cause a salty taste or odor, as well as presenting long-term health effects.[25] The potassium level varied between below detection limit (BDL) to 7.5 mg/L for all water samples except for carbonated waters where the levels were as high as 14.4 mg/L. Only one carbonated water brand exceeded the 12 mg/L level recommended by drinking water guideline standard. Magnesium concentrations ranged from BDL to 5.9, BDL to 7.7, 2.2 to 37.8 and 0.7 to 56.3 mg/L in distilled waters, tap waters, carbonated waters and spring waters respectively. All water brands had magnesium levels falling within the guideline standards except for the two brands of imported spring waters. An average adult ingests as much as 48 mg/L of magnesium daily.

Calcium concentration levels ranged between BDL to 16.1, 0.8 to 21.4, 1.8 to 245.5 and 15.7 to 514.6 mg/L for distilled waters, tap waters, spring waters and carbonated waters, respectively. The majority of the spring waters and carbonated waters exceeded the standard limit of 75 mg/L guidelines. Natural water sources typically contained concentrations of up to 10 mg/L for calcium. However, levels of up to 800 mg/L were found in natural water.[26] The taste threshold for the calcium ion is in the range 100 to 300 mg/L, depending on the associated anion, but higher concentrations are acceptable to consumers. Hardness levels above 500 mg/L are generally considered to be aesthetically unacceptable, although this level is tolerated in some communities.[27] Calcium is one of the major elements responsible for water hardness. Water containing less than 60 mg/L of Ca is considered as soft water. There does not appear to be any convincing evidence that water hardness causes adverse health effects in humans. In contrast, the results of a number of epidemiological studies have suggested that water hardness may protect against disease.[25] Only one sample of each treated tap water and carbonated water showed ammonium levels of 59 and 328 μg/L, respectively.

Anions

Fluoride is an essential element of normal growth and development of humans. Excessive amounts of F^- (4 mg/L) in water supply may result in teeth molting and skeletal

Table 4. Guidelines for the maximum allowable levels of contaminants in drinking water

	Maximum contaminant level guidelines (mg/l) ppm			
Analytes	*IBWA*	*FDA*	*USEPA*	*WHO*
Cations				
Antimony (Sb)	0.006	0.006	0.006	0.02
Aluminum (Al)	0.20	0.20	0.20	0.20
Arsenic (As)	0.01	0.05	0.01	0.01
Barium (Ba)	1	2	2	0.7
Beryllium (Be)	0.004	0.004	0.004	Na
Cadmium (Cd)	0.005	0.005	0.005	0.003
Calcium (Ca)	Na	Na	Na	75
Chromium (Cr) total	0.05	0.1	0.1	0.05
Copper (Cu)	1	1	1.3	2
Iron (Fe)	0.3	0.3	0.3	0.3
Lead (Pb)	0.005	0.005	0.015	0.010
Magnesium (Mg)	Na	Na	50	Na
Manganese (Mn)	0.5	0.5	0.5	0.4
Mercury (Hg)	0.001	0.002	0.002	0.006
Nickel (Ni)	0.1	0.1	Na	0.07
Potassium (K)	Na	Na	Na	12
Selenium (Se)	0.01	0.05	0.05	0.01
Silver (Ag)	0.025	0.1	0.1	0.1
Sodium (Na)	Na	Na	Na	200
Zinc (Zn)	5	5	5	3
Anions				
Chloride (Cl^-)	250	250	250	250
Cyanide (CN^-)	0.1	0.1	0.2	0.07
Fluoride (F^-)	3	3	4	1.5
Nitrite (NO_2^-)	3.3	3.3	3.3	3
Nitrate (NO_3^-)	45	45	45	50
Sulfate ($SO_4^=$)	250	250	250	500
Physical properties				
pH	6.5–8.5	Na	6.5–8.5	6.5–8.5
Total dissolved solids (TDS)	500	500	500	1000
Conductivity (μs/Cm)	Na	Na	Na	400
Microbiological contaminants				
Total coliform/E.coli	None/100 ml	<2.2/100 ml	None	None

International Bottled Water Association (IBWA), Food and Drug Administration (FDA), United States Environmental protection agency (USEPA), World Health Organization (WHO).

fluorosis, whereas low levels (1.0 mg/L) result in diminishing caries reduction.[28] The F^- contents in all of the bottled samples varied between 0.03 and 2.35 mg/L (guideline value is 1.5 mg/L).Chloride (Cl^-) concentrations were 5.5 to 40.8, 0.7 to 28.2, 1.0 to 22.6 and BDL to 29.75 mg/L for carbonated waters, tap waters, distilled waters and spring waters, respectively. No brand had chloride levels that exceeded the standard guideline recommendations. Chloride levels in excess of 250 mg/L can give rise to detectable taste in water, but the threshold depends on the associated cations.[27] Taste thresholds for NaCl and $CaCl_2$ in water are in the range of 200–300 mg/L. Consumption of drinking water containing chloride is not harmful to health. High amounts of chloride can give a salty taste to water. Nitrate (NO_3^-) concentration levels (as NO_3) range from BDL to 50, 4.5–15, 0.17 to 0.39 and 0.09 to 1.1 mg/L in spring waters, carbonated waters, distilled waters and tap waters, respectively. The guidelines standards values for nitrate in drinking water were respected in all of the tested brands except one brand of the spring water samples, which showed a level of 50.04 mg/L, slightly higher than the recommended value of 45 mg/L.

Contamination of drinking water with nitrate presents a health hazard, because nitrate ion can be converted to nitrite ion in the gastrointestinal tract.[29,30] High nitrate levels can cause blue syndrome and certain forms of cancer. The concentration level of nitrite (NO_2^-) in all analyzed samples was much lower than the permissible values of 10 mg/L (3.0 mg/L as Nitrogen) except in one brand of spring water samples showed a level of 10.09 mg/L. Scientific literature suggests that neither nitrate nor nitrite acts directly as a carcinogen in animals, but there is some concern about a possible increased risk of cancer in humans from the endogenous and exogenous formation of N-nitrosamine compounds, many of which are carcinogenic in animals.

The WHO's guideline value for nitrate in drinking water is established solely to prevent methemoglobinemia, which depends on the conversion of nitrate to nitrite. Sulfate (SO_4^{2-}) concentrations in all of the analyzed samples were within the ranges of the WHO and the drinking water standards. Sulfate is generally harmless, except its effect on taste. At levels above 1000 mg/L, there may be laxative effects that can lead to dehydration and gastrointestinal irritation and is of special concern for infants. Reported taste threshold concentrations in drinking water are 250 to 500 mg/L (median 350 mg/L) for sodium sulfate, 250 to1000 mg/L (median 525 mg/L) for calcium sulfate, and 400 to 600 mg/L (median 525 mg/L) for magnesium sulfate.[31] In a survey of 10 to 20 people, the median concentrations of sulfate detectable by taste were 237, 370, and 419 mg/L for the sodium, calcium, and magnesium salts, respectively.[32]

Concentrations of sulfates at which water was determined to have an offensive taste were approximately 1000 and 850 mg/L for calcium and magnesium sulfate, respectively.[35] Addition of calcium sulfate and magnesium sulfate (but not sodium sulfate) to distilled water was found to improve the taste. An optimal taste was found at 270 and 90 mg/L for calcium sulfate and magnesium sulfate, respectively.[29]

Sulfate is one of the least toxic anions. The lethal dose for humans as potassium or zinc sulfate is 45 g.[33,34] The major physiological effects resulting from the ingestion of large quantities of sulfate are catharsis, dehydration, and gastrointestinal irritation. No health-based guideline value for sulfate in drinking water is proposed by either WHO or EPA. However, because of the gastrointestinal effects resulting from the ingestion of drinking water containing high sulfate levels, it is recommended that health authorities be notified of sources of drinking water that contain sulfate concentrations in excess of 500 mg/L. Minor concentrations of Bromide (0.11 mg/L) and phosphates (0.11–0.16) were found in some samples of the carbonated water. Low levels of cyanide were found in treated tap water, carbonated water and distilled water samples, with only one sample of the treated tap water exceeding the MAL of 0.2 mg/L.

Trace metals

Elemental analysis of water samples was carried out using ICPMS as shown in the experimental section. Table 5 shows the mean values of 3 replicates with standard deviation of less than 15% of the mean value. Twenty-seven elements were analyzed with a mass range from 9 mass units for beryllium to 208 mass units for lead. Elements including Boron (B), Silicon (Si) and Strontium (Sr) were found at part per million levels and are reported as mg/L. All other elements are reported as parts per billion (μg/L). Most of the analyzed elements in all of the different brands of bottled water were found to be in concentrations far below the recommended maximum concentration limit guideline recommended by IBWA, FDA EPA and WHO. Few elements were found at levels near or higher than the recommended MCL as follows. One brand of carbonated bottled water (sample # 31) showed elevated molybdenum level of 179.3 ppb, which is much higher than the recommended MCL of 70 μg/L (ppb), two brands of the spring water (sample # 2 and 13) showed a relatively higher level of molybdenum 55.9 and 53.8 μg/L respectively. Levels of molybdenum in drinking water do not usually exceed 10 μg/L, however, in areas near molybdenum mining operations, the molybdenum concentration in finished water can be as high as 200 μg/L. Tap water molybdenum concentrations as high as 580 μg/L have been reported in Colorado.[35]

A provisional guideline value for antimony (Sb) has been set by WHO to a concentration level of 0.005 mg/L. and at 6 ppb (μg/L) by EPA, FDA and IBWA. Although most samples have shown a lower level than the MCL but 3 of the spring water samples, 2 of the treated tap water and 1 of carbonated water showed values above 2 ppb, 1 carbonated water has a value of 3.98 ppb. Most commercially available bottled waters are sold in polyethylene terephthalate (PET) containers. Antimony trioxide is used as a catalyst in the manufacture of PET, which typically contains several hundred milligrams per kilogram (mg/kg) of antimony. The MCL for mercury is 1–2 ppb, most water brands showed levels close to but not exceeding the MCL, value, with one brand of distilled water showing a level of 0.9 ppb.

Relatively high levels of lead was found in only two brands of imported spring water and the other was distilled water with concentration levels of 3.6 and 2.8 ppb, respectively. Lead is a highly toxic metal with no known biological benefit to humans. Its adverse health effects include various cancers, adverse reproductive outcomes, cardiovascular and neurological diseases.[36] No trihalomethanes were detected in any of the samples.

Conclusion

Overall evaluation of the different types of bottled water are shown in Figures 1–3 as an average for all of the analytical measurements that were carried out on all of the 5 different types of bottled waters i.e., spring water fortified tap water, purified tap water, carbonated water and distilled waters. Figure 1 presents the average values for pH and conductivity results. All brands of water showed similar pH values except carbonated water, which was significantly

Table 5. Metal concentration in bottled waters

	Spring Waters															
	Texas (1–5)					*Pennsylvania (6,7)*		*Florida (8,9)*		*Louisiana (10,11)*		*Arkansas (12)*	*Tennessee (13)*	*Ohio (14)*	*Imported (15,16)*	
	Trace Elements Concentration ppb (μg/L)															
Sample #/ Elements	*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*	*11*	*12*	*13*	*14*	*15*	*16*
Be	0.03	0.02	0.01	0.01	0.01	BDL	0.06	BDL	0.06	0.04	0.04	0.03	0.02	0.02	0.02	0.01
Al	BDL	BDL	BDL	BDL	BDL	5.4	6.0	6.5	61.3	4.7	10.3	BDL	BDL	BDL	8.0	BDL
Ti	1.32	1.85	0.91	0.80	0.88	0.67	0.67	0.55	1.48	1.27	1.76	1.13	1.70	1.69	0.60	4.23
V	0.13	BDL	BDL	BDL	BDL	0.18	0.23	1.16	1.34	BDL	BDL	BDL	BDL	BDL	0.05	0.25
Cr	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.21	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Mn	BDL	BDL	1.9	1.9	1.1	BDL	BDL	BDL	0.2	6.0	1.4	BDL	BDL	BDL	BDL	0.2
Fe	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Co	0.06	0.07	0.04	0.04	0.05	0.01	0.04	BDL	0.53	0.27	0.04	0.08	0.05	0.03	0.03	0.14
Ni	1.45	1.74	0.15	0.50	BDL	BDL	BDL	BDL	0.39	1.11	0.62	2.44	1.04	2.11	BDL	3.56
Cu	2.25	BDL	BDL	0.57	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.09	5.25
Zn	0.28	BDL	1.60	1.96	BDL	BDL	BDL	BDL	0.23	0.99	1.40	BDL	BDL	BDL	BDL	1.57
As	0.2	BDL	BDL	BDL	BDL	0.1	0.2	0.4	0.3	BDL	BDL	0.2	BDL	0.2	0.1	0.8
Se	1.62	2.54	0.65	0.64	0.65	0.55	0.82	2.05	0.57	2.13	1.11	1.02	0.78	0.68	0.88	1.28
Mo	4.49	55.90	1.32	0.69	8.18	3.99	5.91	4.24	3.67	7.11	3.72	29.98	53.76	39.74	4.15	5.49
Ag	6.10	BDL	0.10	0.56	0.04	0.09	0.14	0.12	0.17	0.07	0.21	0.16	0.11	0.28	0.11	40.07
Cd	0.24	0.08	0.06	0.10	0.07	0.05	0.16	0.06	0.09	0.11	0.14	0.10	0.06	0.06	0.11	0.89
Sn	BDL	BDL	BDL	BDL	BDL	BDL	0.22	BDL	1.15	0.04	0.05	0.06	BDL	0.07	0.04	0.24
Sb	0.36	0.66	0.11	0.35	0.08	0.18	2.39	0.11	1.99	0.22	0.08	0.53	0.35	0.34	0.49	2.93
Ba	42	66	20	9	6	11	13	2	3	25	21	6	41	3	4	60
Hg	0.53	0.75	0.83	0.58	0.76	0.54	0.75	0.74	0.16	0.76	0.73	0.86	0.55	0.77	0.59	0.34
Tl	0.10	0.07	0.11	0.09	0.06	0.09	0.21	0.13	0.12	0.11	0.12	0.09	0.05	0.10	0.06	0.12
Pb	0.64	0.33	0.51	0.10	0.11	0.22	0.25	0.06	0.72	0.34	0.37	0.38	0.27	0.30	0.17	3.64
	Macro Elements Concentration ppm (mg/L)															
B	5.21	3.49	3.79	4.02	3.73	3.93	3.545	3.55	3.76	3.97	4.77	3.78	3.62	5.42	3.60	11.82
Si	12.4	12.9	10.22	8.58	BDL	7.26	7.18	6.85	7.26	9.75	12.28	8.66	10.89	10.06	6.51	13.59
Sr	0.17	0.20	0.02	0.01	0.01	0.07	0.07	0.10	0.11	0.03	0.03	0.30	0.10	0.29	0.04	0.29

	Purified Municipal Water Fortified With Minerals (17–20)				*Purified Municipal Water Houston, Texas (21–27)*						
	Trace Elements Concentration ppb (μg/L)										
Sample #/Elements	*17*	*18*	*19*	*20*	*21*	*22*	*23*	*24*	*25*	*26*	*27*
Be	BDL	0.09	0.05	0.03	BDL	0.08	0.06	0.04	0.03	0.02	0.02
Al	BDL	0.9	22	13.4	BDL	BDL	BDL	2.9	13.4	BDL	BDL
Ti	0.34	0.36	0.56	0.48	0.43	0.33	0.33	0.71	0.48	0.32	0.93
Mn	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	1.1
Co	BDL	0.04	0.02	BDL	BDL	0.05	0.02	BDL	BDL	0.04	0.06
Ni	BDL	BDL	0.46	BDL	BDL	BDL	BDL	BDL	BDL	0.99	0.06
Cu	0.49	BDL	BDL	BDL	0.26	BDL	BDL	BDL	BDL	BDL	BDL
Zn	BDL	1.05	0.08	BDL	BDL	1.06	BDL	BDL	BDL	0.08	BDL
Se	0.50	0.92	0.42	0.99	0.51	0.25	0.75	0.60	0.99	0.62	0.49
Sr	0.28	0.51	9.28	11.63	0.06	0.11	BDL	BDL	11.63	0.02	3.09
Mo	1.04	12.78	9.6/	5.50	0.47	2.78	0.69	1.32	5.50	9.08	7.87
Ag	0.27	0.41	0.21	0.10	0.15	0.06	0.06	0.25	0.10	0.12	0.05
Cd	0.08	0.09	0.12	0.06	0.06	0.07	0.05	0.06	0.06	0.14	0.05
Sn	BDL	0.43	BDL	BDL	BDL	0.15	0.05	BDL	BDL	BDL	BDL
Sb	0.14	2.19	0.69	0.03	0.11	1.01	0.64	0.28	0.03	0.14	0.08

(*Continued on next page*)

Table 5. *(Continued)*

	Purified Municipal Water Fortified With Minerals (17–20)				*Purified Municipal Water Houston, Texas (21–27)*						
	Trace Elements Concentration ppb (μg/L)										
Sample #/Elements	*17*	*18*	*19*	*20*	*21*	*22*	*23*	*24*	*25*	*26*	*27*
Ba	BDL	BDL	BDL	1	BDL	BDL	BDL	BDL	1	BDL	2
Hg	0.73	0.67	0.55	0.76	0.61	0.47	0.53	0.78	0.76	0.67	0.72
Tl	0.04	0.22	0.12	0.08	0.02	0.17	0.18	0.10	0.08	0.07	0.09
Pb	0.14	0.45	0.45	0.90	0.04	0.59	0.40	0.33	0.90	0.34	0.18
	Macro Elements Concentration ppm (mg/L)										
B	4.47	3.37	3.64	3.95	3.71	3.65	3.66	6.31	3.95	3.65	3.31
Si	3.63	2.81	4.18	4.70	5.51	2.95	3.12	5.50	4.70	3.52	7.72

	Carbonated Water (28–32)					*Distilled Water (33–35)*		
	Trace Elements Concentration ppb (μg/L)							
Sample #/ Elements	*28*	*29*	*30*	*31*	*32*	*33*	*34*	*35*
Be	0.02	0.04	0.03	0.04	0.05	0.05	0.03	0.02
Al	3.3	2.8	102.3	BDL	BDL	5.0	BDL	BDL
P	BDL	BDL	83.5	18.0	BDL	BDL	BDL	BDL
Ti	0.67	0.81	1.54	3.83	1.35	0.51	0.23	0.29
V	BDL	BDL	1.1	BDL	BDL	BDL	BDL	BDL
Cr	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Mn	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Fe	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Co	0.07	0.20	0.98	BDL	0.08	0.02	0.04	BDL
Ni	0.54	3.52	3.03	BDL	0.50	BDL	0.06	0.18
Cu	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Zn	1.29	2.71	5.11	BDL	BDL	BDL	BDL	BDL
As	1	1	BDL	1	BDL	BDL	BDL	BDL
Se	0.81	1.12	0.20	0.40	1.76	0.62	0.41	0.54
Mo	21.14	27.50	2.63	179.30	34.72	11.63	2.23	7.32
Ag	0.11	0.12	0.15	0.21	0.06	0.10	0.07	0.03
Cd	0.11	0.28	0.33	0.16	0.10	0.10	0.10	0.08
Sn	BDL	0.52	1.60	0.11	0.11	BDL	BDL	BDL
Sb	0.96	3.98	2.84	1.66	0.94	0.35	0.09	0.02
Ba	10	8	6	13	34	1	BDL	BDL
Hg	0.19	0.41	0.18	0.61	0.58	0.48	0.91	0.67
Tl	0.06	0.12	0.03	0.16	0.15	0.15	0.08	0.07
Pb	0.05	0.56	0.54	0.59	0.24	2.85	0.28	0.06
	Macro Elements Concentration ppm (mg/L)							
B	3.63	3.47	3.83	10.47	3.58	3.86	2.98	3.57
Si	5.52	5.25	7.02	24.82	9.33	4.33	3.17	3.56
Sr	BDL	BDL	BDL	BDL	BDL	80.10	0.42	0.36

more acidic than all others. On the other hand, conductivity, which reflects the total amount of dissolved solids was found to vary greatly among different types of water. And as expected, carbonated water showed the highest level of conductivity while distilled water showed the least. Anionic species, alkali metals and alkaline earth metals are shown in Figure 2.

Carbonated water showed the highest concentrations among all brands and distilled water brands were the least in their contents. Sulfate was found in the highest concentration among the anions and calcium was the highest among the cations. Trace metals and other macro elements are shown in Figure 3. Spring waters and carbonated waters showed relatively higher concentration of metals than all

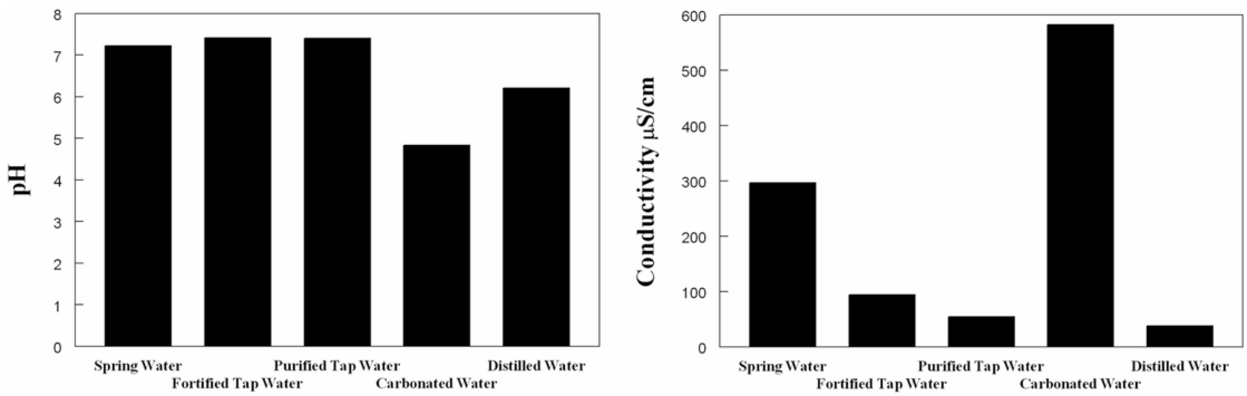

Fig. 1. Average pH and conductivity of analyzed bottled water.

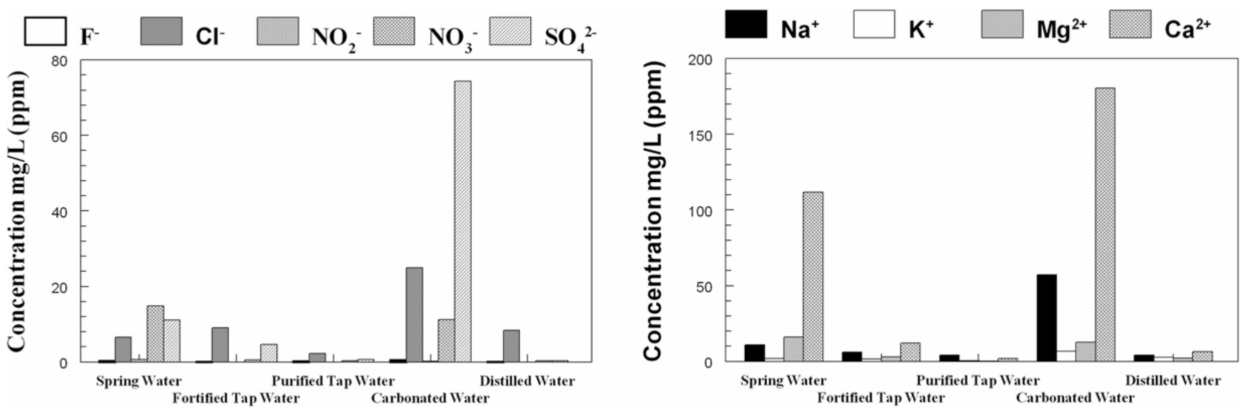

Fig. 2. Average anions and macro elements concentrations.

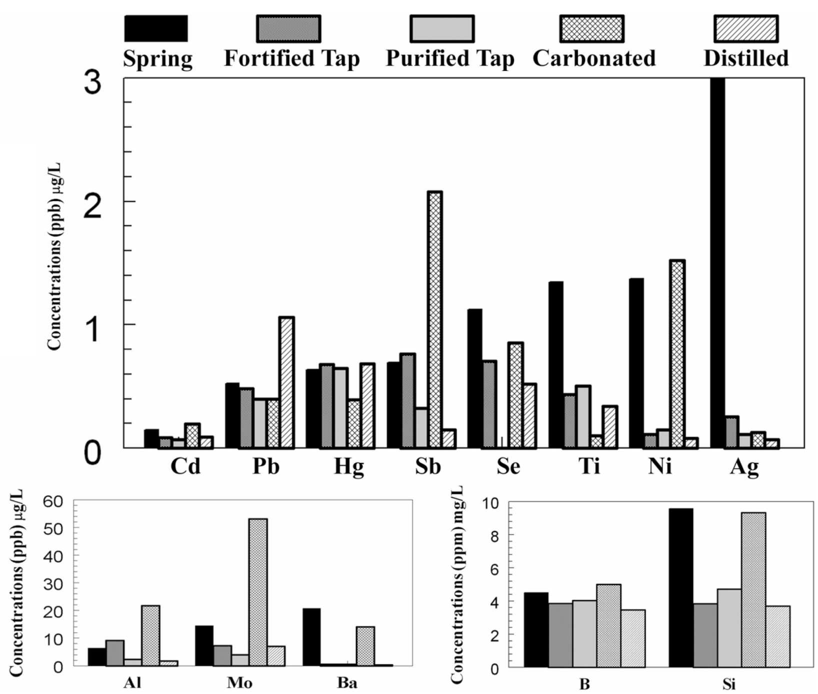

Fig. 3. Average concentration of trace elements in the different types of bottled water.

of the other brands of water. Antimony concentration was shown to be much higher in carbonated water than all other brands of bottled waters. This might be due to the higher acidity of carbonated water that may enhance the leaching of antimony from PET plastic materials of the bottles. Although the majority of the brands of bottled water sold in the greater Houston area comply with guidelines for water quality set nationally or internationally, it does not guarantee higher quality or more safety compared to tap water.

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