FISEVIER

Contents lists available at ScienceDirect

# Journal of Food Composition and Analysis

journal homepage: www.elsevier.com/locate/jfca



# Original Article

# Mineral content of 70 bottled water brands sold on the Turkish market: Assessment of their compliance with current regulations

Cüneyt Güler\*, Musa Alpaslan

Mersin Üniversitesi, Çiftlikköy Kampüsü, Jeoloji Mühendisliği Bölümü, 33343 Mersin, Turkey

#### ARTICLE INFO

Article history: Received 23 April 2008 Received in revised form 10 January 2009 Accepted 2 March 2009

Keywords:
Natural spring water
Bottled water
Water quality
Drinking water standards
Trace elements
ICP-MS
Maximum contaminant level
Turkey
Food safety
Food analysis
Food composition

#### ABSTRACT

In this work, 70 bottled water brands consisting of "natural spring" and "processed drinking" types collected from supermarkets and food stores in eight provinces of Turkey were analyzed for their major and trace element constituents to ascertain their suitability for human consumption. The results obtained were compared with parametric values (PVs) set by European Community Council Directive 98/83/EC and the guideline values (GVs) recommended by World Health Organization (WHO). Al, Co, Cu, Fe, and Mn were non-detectable in most of the samples.  $NH_4^+$ ,  $NO_3^-$ ,  $NO_2^-$ ,  $SO_4^{2-}$ ,  $F^-$ ,  $Cl^-$ , Al, B, Ba, Cd, Cr, Cu, Fe, Mn, Mo, Na, Ni, Pb, Sb, and Se were all below their respective PVs or GVs. However, measured As concentration in one brand was more than three times the standard value set by EC and WHO. Additionally, in six brands, As concentrations were higher than 5  $\mu$ g L<sup>-1</sup>. High concentrations of As in water could pose a risk to individuals who consume those brands on a regular basis.

© 2009 Elsevier Inc. All rights reserved.

# 1. Introduction

Owing to its unique physicochemical properties and its polar structure, water is known as a "universal solvent" and even in pristine environmental conditions it may contain substantial amount of elements in rather high concentrations. At elevated concentrations some elements can be harmful to health (Al Fraij et al., 1999) and can cause morphological abnormalities, mutagenic effects, reduced growth and increased mortality in humans (Nkono and Asubiojo, 1997). Therefore, standards have been developed by national and international organizations to define a quality of water that is safe and acceptable to consumers. Most of these standards set upper limits for physical parameters, chemical constituents and microorganisms that are dangerous, potentially hazardous, or obnoxious to consumers (Magid, 1997).

Freshwater is scarce and resources are unevenly distributed over the world, with much of the water located far from human populations. The total amount of usable freshwater supply is around  $4 \times 10^6 \, \mathrm{km}^3$ , which is only 0.2% of all the water on Earth

(Freeze and Cherry, 1979; Gleick, 1993). It is estimated that 3 billion people will be in the "water scarcity" category (having < 1000 m<sup>3</sup> of renewable water per capita per year) by 2025 (UNEP, 2002). Turkey is currently considered as a state in "water stress" (having 1000–2000 m<sup>3</sup> of renewable water per capita per year). Water-related concerns are most acute in arid or semi-arid areas, and many countries in water-stressed regions rely on alternative or non-conventional water resources (e.g. desalinated seawater, harvested water and treated wastewater). In such countries, consumption of bottled water is a growing practice (Al Fraij et al., 1999; El-Nakhal, 2004). Nowadays, a lot of people living in urban areas are also increasingly consuming bottled water because it is associated with naturalness (Saad et al., 1998) and because they object to unpleasant tastes and odors (e.g. chlorine) from municipal water supplies (Liu and Mou, 2004); it is often regarded as safer than tap water (Armas and Sutherland, 1999). Bottled water is also utilized for consumption in emergency or water shortage situations caused by natural disasters (e.g. drought, earthquake, flood, hurricane, tsunami) or societal disasters (e.g. sabotage, siege, terrorism, war), which can severely damage public and private water supplies for extended periods of time (Güler, 2007a). Today, consumers from all age groups drink bottled water on a daily basis for many different reasons. Therefore, in many parts of the world, bottled water is considered an important

<sup>\*</sup> Corresponding author. Tel.: +90 324 361 0001x7314; fax: +90 324 361 0032. E-mail addresses: cguler@mersin.edu.tr (C. Güler), malpaslan@mersin.edu.tr (M. Alpaslan).

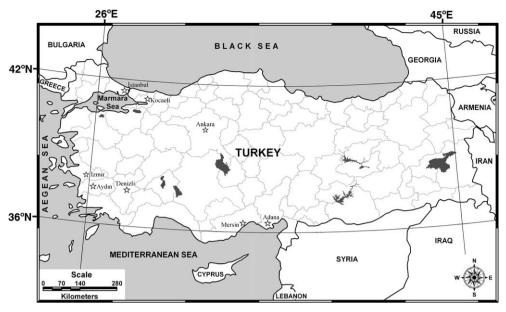


Fig. 1. Geographic locations of the 8 provinces of Turkey, where sampling of 70 different bottled water brands from various commercial establishments was carried out.

element in the human diet and plays a major role in the intake of a number of nutritional and potentially toxic elements (Nkono and Asubiojo, 1997).

In Turkey, groundwater abstracted from drilled wells and water from free-flowing springs are the sources utilized by the bottled water industry. The bottled water industry in Turkey is a very dynamic and competitive market, with key players focused on natural spring water. Smaller companies are mostly century-old and family/municipality-owned, whereas larger firms are under the control of major multinational soft drink companies (Güler, 2007b). As of February 2008, there were 202 domestic brands of bottled water (excluding carbonated and flavored waters) recognized by the Turkish Ministry of Health, which is the sole regulating body of the bottled water industry since 1997. It is estimated that 70% of the households in Turkey regularly drink bottled water, which totaled about  $5.2 \times 10^9 \, \text{L}$  and approximately 78 L per capita in 2002 (Celik, 2003).

Despite the continued expansion of the bottled water industry and positive trend on bottled water consumption in Turkey, reports on bottled water analysis and their mineral contents are scattered and not well documented. The main purpose of this study is to investigate the physical and chemical characteristics of some of the most widely distributed domestic brands of bottled waters in Turkish market. In this study, a total of 70 different brands were sampled, which represents more than 1/3 of the domestic brands currently sold in the market. Additionally, as Turkey moves toward accession to the European Union, this paper aims to assess the compliance of Turkish bottled water brands to several standards around the world including European Community Council Directive 98/83/EC (EC, 1998) and World Health Organization (WHO) guideline values (WHO, 2006).

### 2. Materials and methods

## 2.1. Sample collection and preparation

A total of 70 brands of commercially available, domestically produced bottled waters (all non-carbonated) consisting of "natural spring" and "processed drinking" types were purchased randomly from local supermarkets and independent food stores (locally known as bakkals) in 8 different provinces (Adana, Ankara,

Aydın, Denizli, İstanbul, İzmir, Kocaeli, and Mersin) of Turkey (Fig. 1) between May and November 2007. As indicated on their labels, all the bottled waters had a shelf life of 1 year and were certified by the Turkish Ministry of Health. To keep the brand names anonymous, the waters were given a numerical code from 1 to 70 and this convention was used throughout the study. All bottled waters were in polyethylene terephthalate (PET) containers with plastic screw caps, except brand 59, which was in a transparent glass bottle with an aluminum cap. The holding capacities of bottled water containers ranged from 0.5 to 1.5 L. Sampling of bottled water "from the shelf" was preferred since many urban people tend to consume bottled water and supermarkets and bakkals are the most important sales channels for them. After the collection of samples, they were brought to the laboratory, where they stored in their original bottles under refrigeration at 4 °C and in the dark until analysis. Prior to analysis, manufacturer seals were broken by the analyst in the laboratory and then two 50 mL aliquots were aseptically removed from each bottle for anion and cation analyses. These samples were placed in sterile high density polyethene (HDPE) containers, which were carefully rinsed several times with the sample water. Water samples were not acidified or filtered, since it was our intention to collect and analyze them as "drunk" by the consumer. Collected bottled water samples did not contain any particulates. Manufacturer labels on the bottles were used as a source of basic information on a particular water sample.

# 2.2. Analytical procedures

All analyses were carried out in the Environmental Geochemistry Laboratory at the Mersin University Geological Engineering Department, Mersin, Turkey. The physicochemical parameters of the waters were measured using temperature-compensated WTW Multi 340i/SET (Wissenschaftlich-Technische Werkstätten, Germany) multi-parameter instrument, equipped with electrical conductivity (EC), pH and redox potential (Eh) probes. The probes were calibrated using appropriate standard solutions and procedures before each measurement. The concentrations of ammonia, nitrite, nitrate, sulfate, sulfite, orthophosphate, chloride, and fluoride were determined using Hach Lange DR 2800 spectrophotometer (Hach Lange GmbH, Düsseldorf, Germany) (Table 1).

 Table 1

 Water quality parameters and analytical methods used during the study.

Parameter or analyte	Unit	Analytical method	Instrument
Electrical conductivity (EC)	$\mu S  cm^{-1}$	TetraCon 325 graphite probe	WTW Multi 340i/SET
pH	Standard	Sentix 41-3 glass pH probe	WTW Multi 340i/SET
Redox potential (Eh)	mV	Platinum electrode	WTW Multi 340i/SET
Ammonia (NH <sub>4</sub> <sup>+</sup> )	$ m mgL^{-1}$	Salicylate method	Spectrophotometer <sup>a</sup>
Nitrite (NO <sub>2</sub> <sup>-</sup> )	$mgL^{-1}$	Diazotization method	Spectrophotometer <sup>a</sup>
Nitrate (NO <sub>3</sub> <sup>-</sup> )	$mgL^{-1}$	Cadmium reduction method	Spectrophotometer <sup>a</sup>
Sulfate (SO <sub>4</sub> <sup>2-</sup> )	$mgL^{-1}$	Barium sulfate turbidity	Spectrophotometer <sup>a</sup>
Sulfite (SO <sub>3</sub> <sup>2-</sup> )	$mgL^{-1}$	Colorimetric method	Spectrophotometer <sup>a</sup>
Orthophosphate (PO <sub>4</sub> <sup>3-</sup> )	$mgL^{-1}$	Ascorbic acid method	Spectrophotometer <sup>a</sup>
Chloride (Cl <sup>-</sup> )	$mgL^{-1}$	Mercuric thiocyanate method	Spectrophotometer <sup>a</sup>
Fluoride (F <sup>-</sup> )	$mgL^{-1}$	SPADNS method	Spectrophotometer <sup>a</sup>
Bicarbonate (HCO <sub>3</sub> <sup>-</sup> )	$ m mgL^{-1}$	Titrimetric method (EPA 310.1)	Digital burette
Major ions (Ca, Mg, Na, K, Si)	$ m mgL^{-1}$	Mass spectrometry	ICP-MS (Agilent 7500ce)
Trace elements (Al, As, B, Ba,Br, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb, Se, Sr, Ti, V, Zn)	$\mu \mathrm{g}\mathrm{L}^{-1}$	Mass spectrometry	ICP-MS (Agilent 7500ce)

<sup>&</sup>lt;sup>a</sup> Hach Lange DR 2800.

Bicarbonate was determined in the laboratory by titration according to EPA 310.1. Analyses for total concentrations of 5 major elements (Ca, Mg, Na, K and Si) and 20 trace elements (Al, As, B, Ba, Br, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb, Se, Sr, Ti, V and Zn) were carried out (in separate batches) with inductively coupled plasma-mass spectrometry (ICP-MS). Concentrations of elements in the samples were determined in triplicate by Agilent 7500ce ICP-MS (Agilent Technologies, Tokyo, Japan) equipped with a collusion/reaction cell in the form of Octopole Reaction System (ORS) to minimize polyatomic interferences. The argon gas utilized was of spectral purity (>99.998%). The external standard calibration method was applied to all determinations, using <sup>6</sup>Li, Sc, Ge, Y, In, Tb and Bi internal standard mix. Five-point calibration curves were constructed by analyzing NIST single-element reference standards prepared by serial dilution of stock solutions. The ultrapure water used throughout the period of experimentation had a resistivity of 18 M $\Omega$  cm at room temperature. Deionized water (ELGA Purelab Prima, UK) was used as feed water to produce ultrapure water from the ELGA Purelab UHQ (UK).

## 2.3. Determination of precision and accuracy

The analytical quality control included daily analysis of standards and triplicate analysis of samples and blanks. The accuracy and precision of the analytical technique was evaluated by analyzing a certified standard reference material, CWW-TM-B, Certified Waste Water Trace Metals Solution (B) (High-Purity Standards, Charleston, SC, USA). The relative error (RE, %) is less than  $\pm 5\%$  for all analyzed elements. Our results were in good agreement with the certified values as shown in Table 2. Precision of the instrument was determined by introducing the same quantity of one sample seven times, then the relative standard deviation was calculated (%RSDs are between 0.28 and 1.73). Additionally, after every 10 samples, a standard was analyzed as a sample. If the variation between this sample and standard concentration was more than 10%, the instrument was recalibrated. Additionally, as an independent check of the correctness of the analytical results, the percent charge balance errors (%CBE) were calculated for each bottled water sample as suggested by Freeze and Cherry (1979).

 Table 2

 Accuracy and precision of the ICP-MS analysis results for the certified reference material (CWW-TM-B) and detection limit (DL) for the investigated elements.

•	-		-						,		
Element	Isotope <sup>a</sup>	Measured	values $(n=7)$				%RSD <sup>b</sup>	Certified	Acquisition	$DL^d$	%RE <sup>e</sup>
		Min.	Max.	Median	Mean	S.D.		value	mode <sup>c</sup>		
Al	27	191.90	197.10	195.80	195.31	1.87	0.96	200	No gas	0.056	-2.35
As	75	50.57	52.43	51.33	51.51	0.63	1.22	50	Helium	0.005	3.02
В	10	198.10	204.20	200.80	200.73	2.16	1.08	200	No gas	0.157	0.36
Ba	137	192.40	198.80	195.00	195.13	2.56	1.31	200	No gas	0.010	-2.44
Cd	111	50.44	52.04	51.50	51.30	0.56	1.10	50	No gas	0.004	2.60
Co	59	192.70	198.50	195.90	195.73	2.05	1.05	200	No gas	0.003	-2.14
Cr	52	194.30	201.60	197.80	197.90	2.18	1.10	200	Helium	0.004	-1.05
Cu	63	194.80	202.50	198.30	198.21	2.47	1.25	200	Helium	0.011	-0.89
Fe	56	192.40	203.80	197.20	197.56	3.42	1.73	200	Hydrogen	0.046	-1.22
Mn	55	191.00	196.60	193.70	193.97	2.32	1.20	200	No gas	0.002	-3.02
Mo	95	191.50	199.00	195.40	195.04	2.69	1.38	200	No gas	0.016	-2.48
Ni	60	193.40	201.10	197.60	197.50	2.44	1.24	200	Helium	0.003	-1.25
Pb	208	190.80	198.80	193.30	194.39	2.74	1.41	200	No gas	0.003	-2.81
Sb	121	50.63	52.45	51.88	51.68	0.61	1.17	50	No gas	0.004	3.36
Se	78	51.06	51.94	51.72	51.62	0.28	0.54	50	Hydrogen	0.044	3.24
Sr	88	194.10	199.40	196.30	196.50	1.70	0.87	200	Helium	0.006	-1.75
V	51	196.60	204.40	199.80	199.93	2.34	1.17	200	Helium	0.003	-0.03
Zn	66	208.10	209.80	209.00	208.94	0.58	0.28	200	No gas	0.045	4.47

Concentrations are in  $\mu g L^{-1}$ .

a Isotopic mass of element measured during analysis.

<sup>&</sup>lt;sup>b</sup> Relative standard deviation.

<sup>&</sup>lt;sup>c</sup> Acquisition mode = Octopole reaction system (ORS) acquisition mode.

d Detection limit of the ICP-MS instrument for each isotope.

e Relative error.

Calculated charge balance errors are less than  $\pm 2\%$ , with a mean value of 0.22%.

#### 3. Results and discussion

#### 3.1. Physical parameters and concentration of major ions

It is well established that the chemical composition of natural waters is controlled by many factors that include chemistry of atmospheric precipitation, mineralogy of the rocks encountered along the flowpath, residence time of the groundwater in the aquifer, climate and topography (Güler et al., 2002). Therefore, each bottled water brand has its own characteristic physical and chemical properties that are defined by a unique combination of these factors. In this study, the physicochemical composition of 70 Turkish bottled waters including natural spring (NS) and processed drinking (PD) types were characterized and the results are presented in Table 3. Besides, the standard deviation ( $\pm$ S.D.), mean, median, maximum and minimum parameter values were determined and presented in Table 3, together with parametric values (PVs) set by EC (1998) and the guideline values (GVs) recommended by WHO (2006).

Results show that bottled waters are very different in character and display a wide range of parameter values. The ranges of values for the physical parameters were: 22–662 μSiemens cm<sup>-1</sup> for electrical conductivity (EC); 6.07-9.16 for pH; and -128 to 47 mV for Eh. As it is seen in Table 3, there is a great difference between the EC values of the bottled waters, which is related to the total dissolved solids (TDS) content, the origin of the source, and the treatment or purification method applied during bottling process. There are five natural spring water samples (brands 4, 32, 38, 53, and 68) having very low EC values (<40 µSiemens cm<sup>-1</sup>). A careful examination of major ion composition data for these brands (in Table 3) reveals that concentrations for most of the major ions are below  $1 \text{ mg L}^{-1}$ . As previous studies have shown, long-term consumption of waters low in minerals (e.g. calcium, magnesium and fluoride) have in fact a number of health risks including increased diuresis (WHO, 1980), higher risk of fracture in children (Verd et al., 1992), increased morbidity and mortality from cardiovascular and cerebrovascular diseases (Sauvant and Pepin, 2000; Marque et al., 2003), and increased dental caries, especially in children (Martín et al., 1992). The lack of essential nutrients was also found to be associated with pregnancy disorders or so-called preeclampsia (Melles and Kiss, 1992) and premature and/or lowbirthweight babies (Yang et al., 2002). The concentration range for the major ions were (in mg  $L^{-1}$ ): 0–50.9 for Ca; 0.1–19.0 for Mg; 0.1-76.8 for Na; 0.1-5.3 for K; 0-32.6 for Si; 0.1-0.21 for  $NH_4^+$ ; 0.9-14.2 for  $NO_3^-$ ; 0-0.07 for  $NO_2^-$ ; 0.02-0.55 for  $PO_4^{3-}$ ; 0-62 for  $SO_4^{2-}$ ; 0-0.1 for  $SO_3^{2-}$ ; 0-23.3 for  $Cl^-$ ; 0-0.69 for  $F^-$ ; and 2-189 for HCO<sub>3</sub><sup>-</sup>. The maximum concentrations of Na, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>,  $SO_4^{2-}$ ,  $Cl^-$ , and  $F^-$  were well below their respective PVs or GVs (Table 3). Ca, Mg, K, Si,  $PO_4^{3-}$ ,  $SO_3^{2-}$ , and  $HCO_3^-$  were detected in all the bottled water samples, however, no health-based PVs or GVs were available for these parameters. Nevertheless, epidemiological studies suggest that magnesium may reduce the frequency of sudden death and calcium may help prevent osteoporosis in humans (Garzon and Eisenberg, 1998). In this study, among the samples analyzed, brands 14 and 50 had the highest levels of magnesium (19.0 mg  $L^{-1}$ ) and calcium (50.9 mg  $L^{-1}$ ), respectively.

The results from this study can be used to estimate ingestion amounts of certain elements by consumers. According to Azoulay et al. (2001) adult humans between ages 19 and 50 years require daily 1000 mg Ca<sup>2+</sup>, 310–420 mg Mg<sup>2+</sup> and 2400–3000 mg Na<sup>+</sup>. For the bottled waters examined in this study, adult humans may fulfill only  $\sim\!\!3\%$  of their Ca<sup>2+</sup> dietary reference intake (DRI), between  $\sim\!\!2$  and 1.5% of their Mg<sup>2+</sup> DRI and between only 0.7 and 0.6% of their

Na<sup>+</sup> DRI by drinking 2 L of bottled water per day (calculations were made using mean values). These results indicate that a significant portion of population may be consuming inadequate levels of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , which can have important health-related consequences as reported by Garzon and Eisenberg (1998) and Azoulay et al. (2001). According to results of this study, none of the bottled water brands exceeded the recommended optimal range of  $\text{F}^-$  (0.7–1.2 mg  $\text{L}^{-1}$ ) (Broffitt et al., 2007). It is suggested that consumers should chose to drink bottled water brands with an optimal mineral content, i.e., high levels of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  and low  $\text{Na}^+$  (below 20 mg  $\text{L}^{-1}$ ) to prevent adverse health effects. The pH values of three brands (brands 33, 47, and 53) were acidic (6.47, 6.45, and 6.07, respectively) and values were outside the limits defined by EC Council Directive.

#### 3.2. Concentration of trace elements

Trace element composition of 70 Turkish bottled waters including natural spring (NS) and processed drinking (PD) types are characterized and the results are presented in Table 4. The results obtained are also compared to the PVs set by EC Council Directive (EC, 1998) and GVs recommended by WHO (2006). The concentration range for the 20 trace elements were (in  $\mu g L^{-1}$ ): 0.07-131.1 for Al; 0.12-30.63 for As; 1.3-308.5 for B; 0.3-360.1 for Ba; 0.6-108.5 for Br; 0.29-1.34 for Cd; 0.22-0.57 for Co; 0.14-6.4 for Cr; 0.02-6.78 for Cu; 0.12-48.88 for Fe; 0.01-47.96 for Mn; 0.16-15.33 for Mo; 0.09-7.48 for Ni; 0.21-0.32 for Pb; 0.29-1.23 for Sb; 0.01-0.31 for Se; 0.7-519.3 for Sr; 0.16-5.3 for Ti; 0.01-16.92 for V: and 0.08–364.8 for Zn. Trace elements such as Al. Co. Cu. Fe. Mn. Se. V and Zn were non-detectable in most of the samples. Whereas, Al concentrations in four samples (brands 6, 16, 25, and 70) are relatively high (110.6, 82.1, 131.1, and 73.4  $\mu$ g L<sup>-1</sup>, respectively) but lower than EC PV of 200  $\mu$ g L<sup>-1</sup>. Similarly, Mn concentration in one sample (brand 65) was measured 47.96  $\mu$ g L<sup>-1</sup>; which is very close to EC PV of 50  $\mu$ g L<sup>-1</sup> 1 (Table 4).

The analysis results have shown that in bottled water samples, the maximum concentrations of 13 trace elements (Al, B, Ba, Cd, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb and Se) were all below their respective PVs or GVs (Table 4). However, special attention should be paid to As, since measured arsenic concentration in one sample (brand 61; 30.63  $\mu$ g L<sup>-1</sup>) was more than three times the standard value set by EC (1998) and WHO (2006). Additionally, in five other samples (brands 8, 34, 39, 59, and 62) measured As concentrations were higher than 5  $\mu$ g L<sup>-1</sup>. High concentrations of arsenic in water could pose a risk to individuals who consume those brands on a regular basis. Arsenic is a highly toxic element and chronic arsenic consumption can lead to carcinogenesis (Nordstrom, 2002). Bottled water brands analyzed in this study also contain substantial amounts of Sb, which share some toxicological features with arsenic (Schaumlöffel and Gebel, 1998). Antimony concentrations measured in Turkish bottled waters range between 0.29 and 1.23  $\mu g L^{-1}$  (Table 4). However, these values are well below the EC PV of 5  $\mu g \, L^{-1}$  and WHO GV of 20  $\mu g \, L^{-1}$ . A recent study conducted by Westerhoff et al. (2008) has shown that antimony concentrations in the USA bottled waters range from 0.095 to  $0.521 \mu g L^{-1}$ . They have also shown that antimony concentrations in PET bottles increased over time during storage, especially when they are exposed to elevated temperatures. Antimony can be leached or released from the PET plastic, which is usually made with antimony-based catalysts (Shotyk et al., 2006). In the case of trace elements As and Sb, especially, children may be more at risk due to their greater gastrointestinal absorption and a lower threshold for adverse effects (Cambra and Alonso, 1995). Br, Sr, Ti, V, and Zn were also detected in most of the bottled water samples. However, analytical results for these trace elements cannot be

C. Güler, M. Alpaslan/Journal of Food Composition and Analysis 22 (2009) 728–737

Table 3

Analysis results of physical parameters and major ionic constituents of 70 Turkish bottled waters and their comparison to the EC (1998) and WHO (2006) standards.

Brand code	Type <sup>a</sup>	Physic	ochemical	properties															Discharge <sup>d</sup>	Treatmen
		ECb	pН	Ehc	Ca	Mg	Na	K	Si	NH <sub>4</sub> <sup>+</sup>	NO <sub>3</sub> -	$NO_2^-$	PO <sub>4</sub> <sup>3-</sup>	SO <sub>4</sub> <sup>2-</sup>	SO <sub>3</sub> <sup>2-</sup>	Cl-	F <sup>-</sup>	HCO <sub>3</sub> -		methods <sup>e</sup>
1	PD	71	7.01	5	0.8	3.0	3.8	3.2	0.8	0.20	1.8	0.02	0.04	11.0	0.05	5.1	0.09	9.5		R
!	NS	163	7.54	-37	15.1	2.0	14.3	8.0	21.1	0.14	7.5	0.01	0.36	5.0	0.05	10.2	0.25	60.0	1.0	
1	NS	161	6.77	6	0.3	0.1	0.7	0.2	2.1	0.14	1.8	0.01	0.02	0.0	0.07	0.0	0.01	2.2	2.2	
ļ	NS	39	7.34	-26	4.5	0.7	0.9	0.4	1.9	0.14	1.8	0.01	0.06	4.0	0.03	0.3	0.03	13.0		O + F
5	NS	194	7.07	-10	11.3	3.2	16.7	0.6	8.1	0.13	3.5	0.04	0.14	18.0	0.03	23.3	0.07	31.0	3.5	0
5	NS	84	7.43	-31	6.4	2.8	3.5	0.5	4.1	0.13	8.4	0.01	0.11	12.0	0.05	3.2	0.04	16.0	3.0	
7	NS	51	6.82	4	6.0	0.8	2.2	0.3	4.7	0.14	2.2	0.01	0.09	0.0	0.06	1.3	0.01	25.0	1.0	0
	NS	85	7.11	-13	9.7	1.3	3.7	0.7	8.5	0.13	2.7	0.01	0.08	5.0	0.05	0.9	0.06	38.0	1.0	
)	NS	149	7.99	-63	21.1	1.9	5.2	0.1	5.8	0.11	2.2	0.02	0.04	7.0	0.06	0.8	0.07	78.0	6.0	
10	NS	283	8.18	-73	14.7	5.7	32.5	0.3	9.9	0.13	10.6	0.05	0.06	4.0	0.02	7.0	0.09	130.0	5.0	_
11	NS	161	8.16	-72	22.2	2.6	5.3	0.1	5.6	0.12	3.1	0.03	0.03	8.0	0.03	0.9	0.08	78.0	5.0	0
12	NS	139	7.22	-19	9.8	0.9	17.0	0.1	1.5	0.14	2.2	0.01	0.04	0.0	0.03	0.5	0.03	80.0	2.5	
13	NS	106	7.34	-25	12.5	2.0	4.2	0.5	12.0	0.10	4.9	0.01	0.15	0.0	0.01	1.8	0.11	50.5	1.2	
14	NS	174	9.16	-128	1.7	19.0	1.0	0.1	4.0	0.12	0.9	0.01	0.08	0.0	0.04	1.6	0.04	103.5		
15	NS	184	8.26	-78	28.2	4.1	0.4	0.1	1.2	0.11	2.2	0.01	0.04	1.0	0.04	0.5	0.07	103.0	20.0	0
16	NS	155	7.91	-58	17.9	4.2	6.8	2.3	26.1	0.13	5.8	0.01	0.27	0.0	0.05	2.9	0.19	84.0	3.7	0
17	NS	160	8.08	-68	21.5	2.4	6.3	0.2	6.3	0.14	2.2	0.01	0.06	2.0	0.05	1.1	0.07	86.0		O + F
18	NS	177	8.21	-75	25.4	5.3	0.4	0.1	0.9	0.13	3.1	0.01	0.20	1.0	0.01	0.7	0.06	96.0	1000.0	
19	NS	136	7.91	-58	20.4	1.7	1.6	0.6	7.3	0.13	2.2	0.01	0.08	2.0	0.03	0.9	0.08	72.0	1.0	0
20	NS	191	8.18	-74	28.1	3.4	4.9	0.2	5.6	0.11	2.2	0.01	0.07	6.0	0.02	1.0	0.06	102.0	18.5	
21	NS	188	8.26	-78	28.1	3.4	5.1	0.3	5.6	0.11	2.2	0.03	0.12	12.0	0.02	0.9	0.06	93.5	18.5	
22	NS	207	8.02	-64	32.4	2.7	1.7	1.9	3.1	0.12	2.7	0.01	0.06	7.0	0.03	1.1	0.08	105.0	21.0	0
23	NS	79	6.91	-1	9.7	1.0	4.8	0.4	10.0	0.13	0.9	0.01	0.12	1.0	0.03	1.0	0.10	45.0	4.9	0
24	NS	147	7.85	-55	17.9	3.7	4.2	0.1	8.4	0.14	2.7	0.02	0.11	5.0	0.02	1.2	0.03	75.0	0.8	
25	NS	135	7.48	-34	5.6	1.3	15.7	2.8	26.1	0.14	1.3	0.01	0.06	8.0	0.02	13.1	0.10	38.0		
26	NS	150	8.07	-68	15.6	5.2	4.7	0.2	10.3	0.17	3.1	0.00	0.06	11.0	0.01	2.0	0.07	67.0	1.3	O+F
27	NS	176	8.20	-75	30.6	1.7	0.1	0.1	0.7	0.12	2.7	0.03	0.05	2.0	0.02	0.4	0.03	100.0	1.9	
28	NS	172	8.08	-68	17.1	6.8	6.6	1.6	19.3	0.14	2.2	0.01	0.20	1.0	0.03	0.9	0.12	97.0	6.9	0
29	NS	207	8.12	-71	34.6	8.0	1.1	0.3	1.4	0.13	1.3	0.02	0.04	7.0	0.04	1.7	0.06	130.0	5.0	O+F
30	NS	209	7.60	-41	16.5	6.3	11.0	5.3	32.6	0.18	4.0	0.01	0.55	9.0	0.05	10.6	0.15	84.0	1.6	
31	NS	118	7.88	-50	17.9	2.5	2.1	1.6	4.7	0.12	1.8	0.04	0.06	8.0	0.03	0.7	0.10	60.0		
32	NS	25	7.04	-8	2.1	0.4	1.2	0.4	3.3	0.13	2.7	0.03	0.06	1.0	0.04	0.5	0.04	8.0	2.0	
33	NS	53	6.47	24	8.6	1.3	5.4	0.9	7.1	0.12	3.1	0.01	0.13	0.0	0.02	4.2	0.03	37.0		0
34	NS	144	7.70	-46	16.1	4.5	5.1	3.3	27.3	0.13	6.2	0.01	0.33	0.0	0.00	2.4	0.16	77.0	25.0	
35	NS	322	7.65	-44	3.2	0.3	57.0	0.2	10.7	0.17	0.9	0.01	0.04	62.0	0.02	2.6	0.19	85.0	4.0	
36	NS	200	8.17	-73	33.2	3.2	1.1	0.1	0.8	0.15	1.8	0.01	0.10	4.0	0.04	1.2	0.01	114.0	10.0	
37	NS	194	8.25	-78	33.5	2.4	0.8	0.2	2.1	0.13	2.7	0.01	0.09	0.0	0.08	0.6	0.02	110.0	6.0	
38	NS	22	6.88	1	1.7	0.4	1.4	0.3	3.5	0.14	0.9	0.01	0.08	1.0	0.03	0.6	0.00	9.0	2.0	0
39	NS	221	7.83	-54	17.0	7.2	12.2	3.3	24.6	0.15	10.6	0.01	0.42	15.0	0.02	6.4	0.26	88.0	6.0	
40	NS	198	8.28	-79	30.3	4.0	1.2	0.2	1.8	0.14	2.7	0.01	0.12	1.0	0.10	1.5	0.07	105.0	5.0	
41	NS	251	8.33	-82	17.7	10.2	14.8	2.3	11.1	0.15	7.5	0.01	0.13	8.0	0.07	2.2	0.10	124.5	12.0	
42	NS	89	6.94	-3	7.6	1.2	5.8	1.9	13.6	0.12	2.7	0.01	0.15	15.0	0.06	2.5	0.05	23.0	0.5	
13	NS	276	8.04	-66	35.2	4.3	10.0	0.1	11.6	0.15	2.7	0.02	0.13	5.0	0.01	3.7	0.11	137.0	1.0	
14	NS	106	6.75	8	8.4	1.2	7.1	1.3	7.8	0.12	1.8	0.01	0.04	1.0	0.04	9.9	0.04	31.0	0.6	0
45	NS	239	8.07	-68	24.3	8.1	6.6	1.2	12.3	0.13	14.2	0.01	0.15	3.0	0.01	2.4	0.16	107.0	5.0	
46	NS	72	7.61	-41	9.4	0.9	2.3	0.3	4.2	0.13	2.2	0.01	0.10	3.0	0.06	1.1	0.05	32.5	2.0	
47	NS	109	6.45	25	15.9	1.4	1.7	0.4	6.6	0.16	4.4	0.01	0.07	6.0	0.02	0.6	0.02	46.5	3.0	O + F
48	NS	286	8.21	-75	1.1	0.1	58.8	0.3	3.0	0.14	7.5	0.01	0.13	0.0	0.03	1.1	0.08	148.0		0
49	NS	76	7.59	-40	5.0	1.3	5.8	1.6	9.8	0.13	4.0	0.01	0.25	1.0	0.05	3.6	0.09	27.0	4.6	O + F
50	NS	345	8.17	-73	50.9	9.5	2.5	0.6	2.3	0.13	2.7	0.01	0.07	5.0	0.01	2.7	0.09	189.0	2.0	
51	NS	151	8.08	-68	15.7	5.9	0.7	0.3	2.0	0.15	1.3	0.03	0.08	0.0	0.01	0.5	0.03	75.0	5.0	0

52	NS	70	7.48	-34	4.1	1.4	5.1	1.6	8.4	0.12	2.7	0.01	0.21	9.0	0.04	3.1	0.17	17.0	9.5	0
53	PD	31	6.07	47	0.0	0.1	0.8	0.1	0.0	0.16	0.9	0.01	0.06	0.0	0.06	0.0	0.04	2.0		R
54	NS	64	7.24	-20	6.2	3.2	1.0	0.3	3.1	0.15	2.2	0.01	0.11	0.0	0.04	0.8	0.04	35.0		
55	NS	238	8.06	-67	28.8	6.8	5.2	0.4	3.4	0.15	2.2	0.01	0.08	9.0	0.08	4.1	0.08	111.0	3.0	0
56	NS	112	7.73	-48	16.6	1.9	2.4	0.5	4.3	0.19	2.2	0.04	0.17	9.0	0.05	1.7	0.06	50.5	3.5	
57	NS	164	7.42	-30	7.2	1.2	19.0	2.0	28.2	0.17	0.9	0.01	0.09	1.0	0.04	11.2	0.18	59.5	7.6	
58	NS	227	7.99	-63	31.5	5.2	2.8	2.0	4.6	0.21	1.3	0.01	0.10	37.0	0.03	0.9	0.10	84.0	1.0	
59	NS	342	8.38	-86	45.0	7.1	15.6	4.7	20.2	0.20	3.1	0.01	0.50	20.0	0.06	4.1	0.28	176.0	0.5	0
60	PD	194	7.37	27	6.9	3.5	1.0	0.3	3.3	0.14	3.1	0.01	0.07	5.0	0.05	1.0	0.08	30.0	2.0	R + O + F
61	NS	662	8.93	-115	8.0	1.9	76.8	0.5	10.5	0.15	0.9	0.01	0.06	35.0	0.04	1.4	0.62	188.0	0.2	
62	NS	138	6.93	-2	4.1	1.0	6.4	1.8	11.6	0.13	1.3	0.01	0.47	6.0	0.02	2.9	0.69	20.0	3.0	0
63	NS	361	7.42	-30	9.6	2.1	43.2	1.3	9.7	0.12	3.1	0.01	0.03	1.0	0.02	20.3	0.20	115.0	1.9	O + F
64	NS	540	7.97	-62	11.9	2.6	26.0	0.3	4.9	0.14	1.8	0.06	0.05	11.0	0.01	5.3	0.10	95.0	2.5	O + F
65	NS	278	6.98	-5	6.8	2.4	6.6	0.3	5.0	0.12	1.3	0.01	0.10	1.0	0.05	8.2	0.04	33.0	0.7	0
66	NS	67	7.05	-9	5.6	0.9	3.3	2.0	10.9	0.12	2.2	0.02	0.11	5.0	0.04	1.7	0.08	21.0	13.0	0
67	NS	105	7.83	-54	9.0	0.9	9.5	0.3	5.4	0.13	1.8	0.07	0.10	2.0	0.08	2.0	0.14	50.0	1.0	
68	NS	23	6.96	-4	1.1	0.2	1.6	0.3	0.7	0.12	2.7	0.00	0.07	0.0	0.04	0.3	0.00	6.0	1.5	
69	NS	133	8.10	-70	18.4	1.9	3.7	0.1	5.8	0.20	1.8	0.02	0.10	4.0	0.06	0.9	0.12	65.0	9.5	
70	NS	80	6.88	0	6.6	1.2	5.2	1.6	7.7	0.15	1.3	0.01	0.25	6.0	0.04	4.6	0.14	25.0	1.0	O+F+UV
Minimum		22	6.07	-128	0.0	0.1	0.1	0.1	0.0	0.10	0.9	0.00	0.02	0.0	0.00	0.0	0.00	2.0	0.2	
Maximum		662	9.16	47	50.9	19.0	76.8	5.3	32.6	0.21	14.2	0.07	0.55	62.0	0.10	23.3	0.69	189.0	1000.0	
Median		158	7.78	-49	13.6	2.4	4.9	0.4	5.7	0.13	2.2	0.01	0.09	4.0	0.04	1.6	0.08	73.5	3.0	
Mean		169	7.65	-42	15.3	3.2	8.9	0.9	8.2	0.14	3.1	0.02	0.13	6.4	0.04	3.2	0.10	70.1	21.9	
Standard Dev.		110	0.60	36	11.4	3.1	14.1	1.1	7.5	0.02	2.5	0.01	0.11	9.7	0.02	4.3	0.11	45.1	130.8	
EC Directive		2500	6.5-9.5	-	-	-	200	-	-	0.5	50	0.5	-	250	-	250	1.5	-		
WHO Guidelines		-	-	-	-	-	-	-	-	-	50	0.2	-	-	-	-	1.5	-		

Bold italic numbers indicate values exceeding limits of standards. All concentrations (except as noted below) are in mg L $^{-1}$ .

a Type = NS (natural spring water), PD (processed drinking water).

b EC = electrical conductivity ( $\mu$ Siemens cm $^{-1}$ ).

c Eh = Oxidation-reduction (Redox) potential (mV).

d Spring discharge (L s<sup>-1</sup>).

Treatment methods applied to bottled waters (F: filtration; O: ozonation; R: reverse osmosis; UV: ultraviolet irradiation).

 Table 4

 Trace element concentrations obtained on 70 Turkish bottled waters by ICP-MS analysis and comparison of measured values to the EC (1998) and WHO (2006) standards.

Brand code	Type <sup>a</sup>	Trace ele	Trace elements																		
		Al	As	В	Ва	Br	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Sb	Se	Sr	Ti	V	Zn
1	PD	n.d.	0.20	14.3	4.6	11.5	0.42	n.d.	0.25	n.d.	n.d.	n.d.	1.13	0.15	0.24	0.36	n.d.	9.1	0.31	n.d.	n.d.
2	NS	0.52	0.37	4.2	6.6	60.9	0.30	n.d.	0.34	n.d.	0.85	n.d.	1.22	0.19	0.24	0.37	0.31	233.7	3.39	4.93	2.68
3	NS	n.d.	0.13	2.4	6.6	2.9	0.30	n.d.	0.26	1.66	n.d.	n.d.	0.57	0.40	0.22	0.33	n.d.	2.0	0.52	n.d.	3.03
4	NS	n.d.	0.14	1.9	2.3	3.9	0.30	n.d.	0.22	0.02	0.19	0.54	0.97	0.17	0.22	0.35	n.d.	9.7	0.50	0.04	0.08
5	NS	n.d.	0.50	213.2	27.4	93.5	0.30	n.d.	2.14	n.d.	n.d.	0.31	0.83	7.48	0.23	0.56	0.14	51.9	1.52	0.44	2.49
6	NS	110.60	0.27	14.6	8.3	30.2	0.29	n.d.	0.50	0.39	48.88	0.64	0.49	0.32	0.26	0.35	0.31	37.4	2.30	0.12	1.97
7	NS	n.d.	0.29	3.7	7.4	13.6	0.30	n.d.	0.22	n.d.	n.d.	1.57	0.63	1.08	0.22	0.38	n.d.	31.2	0.93	n.d.	0.41
8	NS	n.d.	7.42	2.9	1.6	11.5	0.34	n.d.	0.54	n.d.	n.d.	0.10	2.14	0.57	0.22	0.33	n.d.	61.2	1.43	0.06	3.22
9	NS	0.88	0.46	9.2	5.3	9.3	0.29	n.d.	0.60	n.d.	n.d.	n.d.	0.87	0.15	0.22	0.38	0.19	66.8	1.07	5.68	n.d.
10	NS	n.d.	0.44	18.4	1.9	52.0	0.29	n.d.	6.40	n.d.	n.d.	n.d.	0.78	0.14	0.22	0.38	0.22	27.1	1.69	2.75	n.d.
11	NS	n.d.	0.66	9.2	18.4	8.6	0.29	n.d.	0.63	n.d.	n.d.	0.14	0.89	0.13	0.22	0.38	0.18	77.3	1.06	5.25	101.
12	NS	n.d.	0.17	3.5	3.9	4.6	0.29	n.d.	0.44	n.d.	n.d.	n.d.	0.43	0.18	0.22	0.35	n.d.	30.4	0.40	n.d.	3.66
13	NS	n.d.	0.28	2.7	0.8	14.3	0.29	n.d.	0.44	n.d.	n.d.	1.66	0.60	0.14	0.22	0.34	n.d.	43.9	2.09	1.68	0.71
14	NS	n.d.	0.13	2.1	0.6	10.1	0.29	n.d.	2.37	n.d.	n.d.	n.d.	0.49	0.12	0.22	0.39	n.d.	6.7	0.79	n.d.	n.d.
15	NS	n.d.	0.52	5.0	3.9	6.5	0.29	n.d.	0.63	n.d.	n.d.	n.d.	0.56	0.16	0.22	0.35	0.01	30.9	0.36	0.39	0.50
16	NS	82.11	1.28	13.3	45.0	27.3	0.29	n.d.	0.34	n.d.	20.97	0.04	0.80	0.13	0.23	0.44	0.02	225.1	4.92	5.74	n.d.
17	NS	0.13	0.39	12.4	3.4	8.1	0.29	n.d.	0.81	n.d.	n.d.	n.d.	0.68	0.09	0.21	0.35	0.18	69.4	1.15	9.71	n.d.
18	NS	n.d.	0.98	5.0	7.1	6.2	0.29	n.d.	0.64	n.d.	n.d.	n.d.	1.84	0.14	0.21	0.41	0.14	52.6	0.31	0.18	0.24
19	NS	n.d.	0.57	3.0	2.3	8.4	0.30	n.d.	0.54	0.03	n.d.	0.47	3.17	0.21	0.21	0.39	0.05	38.7	1.36	0.33	0.89
20	NS	n.d.	0.63	8.8	18.9	7.1	0.29	n.d.	0.54	n.d.	n.d.	n.d.	0.73	0.11	0.21	0.37	0.05	103.3	1.07	3.68	n.d.
21	NS	n.d.	0.03	8.6	17.2	6.6	0.29	n.d.	0.58	n.d.	n.d.	n.d.	0.75	0.11	0.21	0.39	0.15	103.5	1.02	3.69	n.d.
22	NS	n.d.	0.71	3.6	18.3	4.5	0.29	n.d.	0.37	n.d.	n.d.	n.d.	1.97	0.10	0.21	0.39	0.10	53.5	0.67	0.90	n.d.
	NS			2.3			0.29							0.12	0.21	0.39		92.5			25.2
23		n.d.	4.55		2.2	10.6		n.d.	0.21	2.57	n.d.	n.d.	0.68				n.d.		1.76	n.d.	
24	NS NC	n.d.	0.43	12.2	0.9	8.6	0.29	n.d.	0.81	n.d.	n.d.	n.d.	0.36	0.11	0.21	0.52	0.07	33.5	1.52	7.58	0.48
25	NS	131.10	1.51	30.3	1.2	70.1	0.29	n.d.	0.61	n.d.	35.57	0.56	0.56	0.22	0.31	0.43	0.07	21.4	5.30	0.13	0.78
26	NS	n.d.	0.17	4.7	0.9	12.0	0.29	n.d.	0.69	n.d.	n.d.	0.01	0.38	0.13	0.22	0.36	0.28	52.2	1.67	6.36	0.53
27	NS	2.37	0.29	1.6	3.6	4.6	0.29	n.d.	0.49	n.d.	0.21	n.d.	0.26	0.14	0.21	0.35	0.01	19.5	0.31	0.21	n.d.
28	NS	n.d.	0.73	5.2	3.9	13.3	0.29	n.d.	1.01	n.d.	n.d.	n.d.	0.41	0.15	0.22	0.33	n.d.	150.5	3.06	5.05	80.93
29	NS	n.d.	0.31	3.7	14.2	11.3	0.29	n.d.	0.42	n.d.	n.d.	n.d.	1.63	0.19	0.21	0.37	0.08	519.3	0.40	1.24	1.07
30	NS	5.32	2.21	22.3	23.0	58.5	0.29	n.d.	0.76	n.d.	2.74	0.06	0.43	0.17	0.23	0.43	0.14	145.6	4.79	4.56	1.59
31	NS	0.07	0.23	2.2	13.8	6.2	0.30	n.d.	0.31	n.d.	0.21	n.d.	3.21	0.15	0.21	0.35	0.05	76.9	0.82	0.47	0.38
32	NS	2.06	0.15	1.3	3.4	3.3	0.50	n.d.	0.17	n.d.	0.88	0.10	0.43	0.16	0.21	0.33	n.d.	13.3	0.71	n.d.	3.97
33	NS	n.d.	0.52	3.7	360.1	25.5	0.30	n.d.	0.23	5.51	n.d.	2.74	0.20	0.52	0.22	0.38	n.d.	22.4	0.82	0.03	4.06
34	NS	0.07	9.90	23.5	35.7	21.1	0.29	n.d.	1.18	n.d.	n.d.	n.d.	1.47	0.17	0.21	0.65	0.11	202.8	3.94	6.91	n.d.
35	NS	n.d.	3.52	308.5	1.8	15.2	0.29	n.d.	0.18	n.d.	n.d.	n.d.	2.80	0.10	0.21	0.48	n.d.	21.3	1.61	0.01	n.d.
36	NS	2.00	4.00	16.1	13.3	8.4	0.29	n.d.	0.26	n.d.	n.d.	n.d.	0.40	0.12	0.21	0.54	0.01	62.5	0.31	0.09	n.d.
37	NS	0.14	0.28	4.3	6.0	6.6	0.29	n.d.	0.99	n.d.	n.d.	n.d.	0.27	0.10	0.21	0.96	0.09	41.3	0.52	1.29	n.d.
38	NS	n.d.	0.17	2.3	3.1	5.4	0.29	n.d.	0.34	n.d.	n.d.	0.04	0.21	0.16	0.21	0.36	n.d.	11.6	0.74	n.d.	0.19
39	NS	n.d.	6.73	40.0	10.4	31.6	0.29	n.d.	1.15	n.d.	n.d.	0.36	2.47	0.14	0.22	0.41	0.07	131.9	3.70	16.92	n.d.
40	NS	n.d.	0.20	6.3	3.7	9.7	0.29	n.d.	0.70	n.d.	n.d.	n.d.	0.27	0.13	0.21	0.35	n.d.	76.5	0.49	0.65	n.d.
41	NS	1.12	0.61	8.0	0.8	21.8	0.29	n.d.	3.12	n.d.	n.d.	n.d.	1.51	0.13	0.22	0.34	0.09	136.4	1.99	14.07	n.d.
42	NS	0.22	2.49	26.1	37.4	27.5	0.29	n.d.	0.24	n.d.	0.43	0.05	0.29	0.90	0.22	0.92	n.d.	51.9	2.25	0.29	1.42
43	NS	1.29	0.32	7.7	0.4	18.9	0.29	n.d.	0.19	n.d.	0.12	n.d.	0.31	0.11	0.22	0.38	0.09	54.9	1.99	5.60	n.d.
44	NS	n.d.	0.17	3.4	49.7	61.0	0.33	n.d.	0.42	n.d.	n.d.	1.41	0.18	3.81	0.22	0.71	0.08	34.7	1.35	0.04	8.45
45	NS	n.d.	0.37	6.7	0.9	33.4	0.29	n.d.	1.13	n.d.	n.d.	n.d.	0.68	0.15	0.21	0.37	0.09	154.0	2.05	7.09	n.d.
46	NS	n.d.	0.43	2.9	5.2	16.4	0.30	n.d.	0.22	6.78	n.d.	0.23	0.21	1.69	0.23	0.38	n.d.	63.7	0.85	n.d.	11.9
47	NS	4.62	0.43	11.8	4.7	4.2	0.29	n.d.	0.22	n.d.	0.12	0.23	0.21	0.13	0.23	0.38	0.04	48.0	1.21	0.14	n.d.
.,	140	16.88	0.50	5.7	0.7	10.1	0.29	n.d.	0.46	11.0.	8.10	0.10	0.27	0.15	0.24	0.37	0.04	2.5	0.93	0.62	0.73

49	NS	n.d.	1.55	3.8	11.9	31.3	0.30	n.d.	0.28	n.d.	n.d.	0.40	0.24	0.48	0.22	0.34	0.03	34.5	1.78	0.12	0.57
50	NS	0.77	0.97	15.6	16.1	15.9	0.29	n.d.	0.50	n.d.	n.d.	n.d.	0.95	0.12	0.21	0.41	0.07	97.0	0.57	0.70	n.d.
51	NS	n.d.	0.15	1.4	3.7	4.0	0.29	n.d.	0.23	n.d.	n.d.	n.d.	0.64	0.10	0.21	0.35	n.d.	14.2	0.51	0.27	n.d.
52	NS	n.d.	2.74	5.8	2.6	22.9	0.30	n.d.	0.18	n.d.	n.d.	0.04	0.70	0.56	0.21	0.42	0.04	23.2	1.55	n.d.	0.23
53	PD	n.d.	0.12	4.8	0.3	0.7	0.29	n.d.	0.26	n.d.	n.d.	n.d.	0.17	0.17	0.21	0.34	n.d.	0.7	0.16	n.d.	n.d.
54	NS	n.d.	0.90	21.5	1.8	3.3	0.29	n.d.	0.16	n.d.	n.d.	n.d.	0.30	0.22	0.21	0.35	n.d.	40.1	0.68	1.14	1.45
55	NS	0.40	0.51	62.8	28.1	18.9	0.29	n.d.	0.65	n.d.	n.d.	n.d.	1.72	0.11	0.22	0.38	0.05	129.3	0.74	0.87	n.d.
56	NS	0.11	0.91	3.3	7.5	16.7	0.29	n.d.	0.25	n.d.	n.d.	n.d.	0.30	0.62	0.21	0.43	0.02	50.9	0.85	n.d.	0.39
57	NS	5.06	4.93	47.6	0.9	54.9	0.29	n.d.	0.23	n.d.	1.32	n.d.	1.22	0.11	0.22	0.52	0.09	18.8	4.19	0.05	n.d.
58	NS	n.d.	0.43	3.1	20.5	7.7	0.31	n.d.	0.31	n.d.	n.d.	1.03	15.33	0.15	0.21	0.44	0.13	207.9	0.88	0.63	n.d.
59	NS	n.d.	5.09	70.9	85.0	23.1	0.30	n.d.	0.24	n.d.	n.d.	n.d.	1.23	0.24	0.32	0.58	0.14	432.7	3.36	2.22	5.53
60	PD	n.d.	1.05	22.7	2.2	3.8	0.32	n.d.	0.17	n.d.	0.64	n.d.	0.35	0.14	0.23	0.29	n.d.	45.2	0.72	1.27	1.03
61	NS	2.03	30.63	268.3	3.7	15.8	0.29	n.d.	0.26	n.d.	n.d.	0.08	6.58	0.11	0.22	1.23	n.d.	137.2	1.76	1.02	n.d.
62	NS	7.55	8.48	9.8	1.5	22.4	0.32	0.22	0.16	0.37	n.d.	0.24	1.08	1.05	0.22	0.58	0.06	16.8	2.13	n.d.	1.11
63	NS	0.84	0.37	75.1	49.6	108.5	0.31	n.d.	1.09	n.d.	n.d.	3.38	0.65	4.72	0.22	1.06	0.03	76.9	1.68	0.17	16.43
64	NS	n.d.	0.18	6.7	1.5	13.2	1.34	n.d.	0.25	n.d.	n.d.	n.d.	0.25	0.23	0.22	0.70	0.17	48.2	0.96	0.23	0.77
65	NS	n.d.	0.22	6.0	18.3	48.2	1.15	0.57	0.20	0.62	0.17	47.96	0.23	1.55	0.22	0.93	0.18	34.8	1.00	n.d.	364.80
66	NS	n.d.	1.49	8.8	31.4	28.4	0.34	n.d.	0.42	2.13	n.d.	0.26	0.22	1.57	0.23	0.52	0.02	36.7	1.92	0.04	7.27
67	NS	n.d.	0.39	34.1	8.2	11.7	0.64	n.d.	0.17	n.d.	n.d.	0.01	0.24	0.16	0.21	1.01	0.03	29.4	1.03	0.97	1.20
68	NS	n.d.	0.14	43.3	0.7	1.4	0.34	n.d.	0.14	0.93	n.d.	0.01	0.16	0.13	0.22	1.15	n.d.	3.6	0.26	n.d.	3.18
69	NS	1.62	0.35	4.6	0.6	7.9	0.29	n.d.	1.01	n.d.	0.18	n.d.	0.37	0.10	0.21	0.34	0.06	47.5	1.18	9.65	0.24
70	NS	73.36	0.43	4.8	42.4	27.1	0.30	n.d.	0.37	n.d.	32.77	1.51	0.23	0.18	0.24	0.34	0.05	75.2	3.37	0.77	3.23
Minimum		0.07	0.12	1.3	0.3	0.6	0.29	0.22	0.14	0.02	0.12	0.01	0.16	0.09	0.21	0.29	0.01	0.7	0.16	0.01	0.08
Maximum		131.10	30.63	308.5	360.1	108.5	1.34	0.57	6.40	6.78	48.88	47.96	15.33	7.48	0.32	1.23	0.31	519.3	5.30	16.92	364.80
Median		1.62	0.44	6.5	4.7	11.8	0.29	0.40	0.40	0.93	0.75	0.28	0.58	0.15	0.22	0.38	0.08	48.1	1.07	0.87	1.42
Mean		16.79	1.71	23.6	16.4	20.1	0.33	0.40	0.63	1.91	8.57	2.08	1.08	0.51	0.22	0.47	0.10	74.4	1.50	2.64	15.58
Standard Dev.		36.36	4.07	54.1	44.4	21.2	0.17	0.25	0.87	2.27	15.19	8.41	1.99	1.13	0.02	0.21	0.08	88.4	1.20	3.69	57.89
EC Directive		200	10	1000	_	_	5	_	50	2000	200	50	70	20	10	5	10	-	_	_	_
WHO Guidelines		-	10	500	700	-	3	-	50	2000	-	400	70	70	10	20	10	-	-	-	-

n.d. = not detected or values below ICP-MS detection limits given in Table 2. All concentrations are in  $\mu$ g L<sup>-1</sup>. <sup>a</sup> NS = natural spring water, PD = processed drinking water.

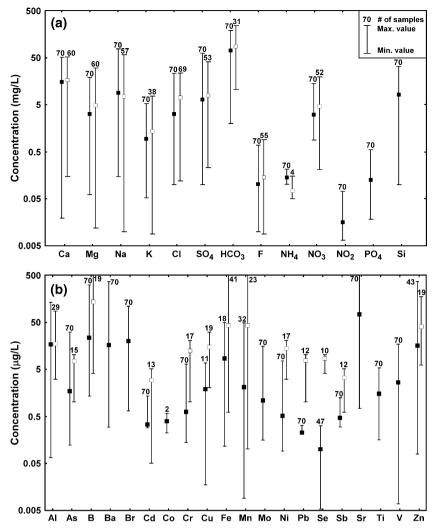


Fig. 2. Graphical comparison of the study results with the values reported on the bottled water labels. Whiskers in the graphs indicate concentration range (min.—max.) for the parameters. Filled and empty boxes indicate mean concentration values obtained by this study (■) and values reported on the bottled water labels (□), respectively.

evaluated, since no health-based PVs or GVs were determined for those elements by both EC (1998) and WHO (2006).

# 3.3. Comparison of analytical results with the reported label values

Comparison of the study results with the reported label values for (a) major species (Ca, Mg, Na, K, Si, NH<sub>4</sub><sup>+</sup>, Cl<sup>-</sup>, HCO<sub>3</sub><sup>-</sup>, F<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, PO<sub>4</sub><sup>3-</sup>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>) and (b) trace elements (Al, As, B, Ba, Br, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb, Se, Sr, Ti, V, Zn) were made in Fig. 2. As it is seen in this figure, no concentration values were reported on the bottle labels for the parameters NO<sub>2</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, Si, Ba, Br, Co, Mo, Sr, Ti and V. Hence, for these parameters no comparison can be made. Mean concentrations of analytical and labeled values are relatively close for the major species, but they are generally lower than the values reported on the bottle labels (Fig. 2a). Nevertheless, a similar trend was not observed for the trace elements (Fig. 2b) and comparison of analytical and labeled values reveals no relationship between the two. It should be noted that the number of reported label values is much lower than (in some cases missing) the number of analytical results for a long range of major and trace elements, which significantly hinders comparison of the results. However, statistically significant (at p < 0.05 level) but low correlations (r) exist between the analytical and labeled values of pH (0.65), Ca (0.69), Mg (0.68), Na (0.59), K (0.65), B (0.68), Cl<sup>-</sup> (0.76),  $SO_4^{\ 2-}$  (0.68),  $F^-$  (0.56), and  $NO_3^-$  (0.35). For the rest of the

parameters, especially for the trace elements, no significant correlations were present. These results may be due to the fact that the waters are subjected to several processes before bottling (e.g. filtration, ozonation, purification, ultraviolet irradiation, removal and/or addition of certain elements/minerals). Most of the time, these processes are not indicated on the bottle labels (Table 3). Additionally, information reported on the bottle labels is simply based on analysis results obtained several decades ago, during which significant changes may have occurred in the source water chemical composition. The abnormal differences between measured and labeled values can be due to simply a typographic error on labels (e.g. wrong decimal places or reporting units). Furthermore, additional changes to water chemistries can occur during the storage of the bottles such as (co)precipitation of constituents or leaching of some constituents from the bottle, as well as from production line of the bottling plant itself. Another reason for the observed changes may be due to accuracy, precision or detection limits of the employed analytical methods/instruments used. Whatever the reason for the change, the results have shown that there can be a significant difference between labeled and measured values, which is even more pronounced for the trace element data. It is believed that with this study, for the first time, a synoptic survey of the Turkish bottled waters was conducted using a unified methodology (e.g. analysis were made using the same methods and instruments).

#### 4. Conclusions

In this work, a survey of the major and trace element content of 70 bottled water brands currently sold in Turkish market was carried out. The physicochemical quality (physical parameters, major constituents and trace elements) of the brands studied was extremely variable, which possibly depends on many factors such as; natural environment (geological setting, climate, topography, etc.), source water composition and type of treatment/purification technique(s) applied during the production. Additional changes in the water chemistries may also occur during storage and transportation, especially when bottles are exposed to direct sunlight. Of the 70 brands of bottled water studied, a total of 4 water treatment/ purification techniques (filtration, reverse osmosis, ozonation, and ultraviolet irradiation) were used by the various brands as indicated on their labels. Al, Co, Cu, and Fe were non-detectable in most of the samples. Some bottled water samples had low pH values and high concentrations of non-essential elements (e.g. As) exceeding the EC and WHO drinking water standards. These bottled water brands may not be suitable for human consumption. As shown by previous studies arsenic can cause chronic or acute poisoning and should be eliminated from drinking water. Other elements detected were all below the PVs established by the EC and were within the WHO GVs. Mn concentration in one brand was measured 47.96  $\mu$ g L<sup>-1</sup>; which is very close to EC PV of  $50 \mu g L^{-1}$ . On the contrary several brands had very low electrical conductance (EC) values (<40 µSiemens cm<sup>-1</sup>) and major ion concentrations (below  $1 \text{ mg L}^{-1}$ ). These waters are either naturally low in TDS or artificially stripped of their mineral contents. As previous studies have shown, long-term consumption of waters low in minerals (e.g. calcium, magnesium and fluoride) may be responsible for important health problems. Therefore, new international water quality standards determining the minimum acceptable limits for these compounds are needed to protect public health. Additionally, no standards have been established by both EC and WHO for the following parameters analyzed during this study: Eh, Ca, Mg, K, Si, PO<sub>4</sub><sup>-3</sup>, SO<sub>3</sub><sup>-2</sup>, HCO<sub>3</sub><sup>-</sup>, Br, Co, Sr, Ti, V, and Zn.

The number and type of parameters reported on the labels of Turkish bottled water showed a lack of homogeneity. Basic parameters (major ions) were usually indicated, whereas, for example, none of the brands reported concentration values of  $NO_2^-$ ,  $PO_4^{3-}$ , Si, Ba, Br, Co, Mo, Sr, Ti or V on the bottle labels. On the whole, the concentration of species measured in this study was slightly lower than the values reported on the labels. This study has also shown that bottled water is not necessarily safer than tap water, and consumers should be aware of this fact. Additionally, in light of these findings and recent concerns about arsenic in some provinces of Turkey (e.g. İzmir and Ankara), there is a need for a nationwide survey about the quality of municipal waters (e.g. tap water) as well.

# Acknowledgements

The authors gratefully acknowledge the energetic and enthusiastic help of many relatives, friends, colleagues, and students, without whom the collection of all 70 bottled water brands would have been impossible.

#### References

- Al Fraij, K.M., Abd El Aleem, M.K., Al Ajmy, H., 1999. Comparative study of potable and mineral waters available in the State of Kuwait. Desalination 123, 253–264.Armas, A.B., Sutherland, J.P., 1999. A survey of the microbiological quality of bottled water sold in the UK and changes occurring during storage. International Journal of Food Microbiology 48, 59–65.
- Azoulay, A., Garzon, P., Eisenberg, M.J., 2001. Comparison of the mineral content of tap water and bottled waters. Journal of General Internal Medicine 16, 168–175.
- Broffitt, B., Steven, M.L., Warren, J.J., Cavanaugh, J.E., 2007. An investigation of bottled water use and caries in the mixed dentition. Journal of Public Health Dentistry 67, 151–158.
- Cambra, K., Alonso, E., 1995. Blood lead levels in 2- to 3-year-old children in the Greater Bilbao area (Basque county, Spain): relation to dust and water levels. Archives of Environmental Health 50, 362–366.
- Çelik, P., 2003. Milliyet Business: İlk sucumuz Hamidiye'ydi. Retrieved 2003-09-10: http://www.milliyet.com.tr/2003/09/01/business/bus06.html#
- EC, 1998. Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption. Official Journal of the European Communities L 330. 32–54.
- El-Nakhal, H.A., 2004. Alternatives to tap water: a case study of the Gaza Strip, Palestine. Environmental Geology 46, 851–856.
- Freeze, R.A., Cherry, J.A., 1979. Groundwater. Prentice-Hall, Englewood Cliffs, NJ. Garzon, P., Eisenberg, M.J., 1998. Variation in the mineral content of commercially available bottled waters: implications for health and disease. American Journal of Medicine 105. 125–130.
- Gleick, P.H., 1993. Water in Crisis: A Guide to the World's Freshwater Resources. Oxford University Press, New York.
- Güler, C., Thyne, G.D., McCray, J.E., Turner, A.K., 2002. Evaluation of graphical and multivariate statistical methods for classification of water chemistry data. Hydrogeology Journal 10, 455–474.
- Güler, C., 2007a. Evaluation of maximum contaminant levels in Turkish bottled drinking waters utilizing parameters reported on manufacturer's labeling and government-issued production licenses. Journal of Food Composition and Analysis 20, 262–272.
- Güler, C., 2007b. Characterization of Turkish bottled waters using pattern recognition methods. Chemometrics and Intelligent Lab Sys 86, 86–94.
- Liu, Y., Mou, S., 2004. Determination of bromate and chlorinated haloacetic acids in bottled drinking water with chromatographic methods. Chemosphere 55, 1253–1258
- Magid, H.M.A., 1997. Assessment of drinking water quality in the Al-Gassim region of Saudi Arabia. Environment International 23, 247–251.
- Martín, M., Hardisson, A., Alvarez, R., 1992. Fluoride concentration in beers and soft drinks. Journal of Food Composition and Analysis 5, 172–180.
- Marque, S., Jacqmin-Gadda, H., Dartigues, J.F., Commenges, D., 2003. Cardiovascular mortality and calcium and magnesium in drinking water: an ecological study in elderly people. European Journal of Epidemiology 18, 305–309.
- Melles, Z., Kiss, A.S., 1992. Influence of the magnesium content of drinking water and of magnesium therapy on the occurrence of preeclampsia. Magnesium Research 5, 277–279.
- Nkono, N.A., Asubiojo, O.I., 1997. Trace elements in bottled and soft drinks in Nigeria—a preliminary study. Science of the Total Environment 208, 161–163. Nordstrom, D.K., 2002. Worldwide occurrences of arsenic in ground water. Science 296 2143–2145
- Saad, B., Pok, F.W., Sujari, A.N.A., Saleh, M.I., 1998. Analysis of anions and cations in drinking water samples by capillary ion analysis. Food Chemistry 61, 249–254.
- Sauvant, M.-P., Pepin, D., 2000. Geographic variation of the mortality from cardiovascular disease and drinking water in a French small area (Puy de Dome). Environmental Research 84, 219–227.
- Schaumlöffel, N., Gebel, T., 1998. Heterogeneity of the DNA damage provoked by antimony and arsenic. Mutagenesis 13, 281–286.
- Shotyk, W., Krachler, M., Chen, B., 2006. Contamination of Canadian and European bottled waters with antimony from PET containers. Journal of Environmental Monitoring 8, 288–292.
- UNEP, 2002. Global Environment Outlook 3—Past, Present and Future Perspectives. Earthscan Publications Ltd., London.
- Verd, V.S., Domínguez, S.J., González, Q.M., Vidal, M.M., Mariano, S.A.C., de Roque, C.C., Sevilla, M.J.M., 1992. Association between calcium content of drinking water and fractures in children (in Spanish). Anales Españoles de Pediatría 37, 461–465.
- Westerhoff, P., Prapaipong, P., Shock, E., Hillaireau, A., 2008. Antimony leaching from polyethylene terephthalate (PET) plastic used for bottled drinking water. Water Resources 42. 551–556.
- WHO, 1980. Guidelines on Health Aspects of Water Desalination. ETS/80.4. World Health Organization, Geneva.
- WHO, 2006. Guidelines for drinking-water quality, first addendum to third edition. Recommendations, vol. 1. WHO, Geneva.
- Yang, Ch-Y., Chiu, H.F., Chang, Ch.-Ch., Wu, T.N., Sung, F.-Ch., 2002. Association of very low birth weight with calcium levels in drinking water. Environmental Research Section A 89, 189–194.