

ELEMENTAL VARIATION ON BOTTLED DRINKING WATER

O. AL-DAYEL*, J. HEFNE, S. BALAWEE and T. AL-AJYAN

King Abdulaziz City for Science and Technology, P.O. Box 6086, RIYADH-11442, SAUDI ARABIA

ABSTRACT

The concentration of some inorganic metals is restrictive factor for drinking water quality, generally as consequence of their effects on the health. This fact makes necessary its regulation and monitoring. Inductively Coupled Plasma Mass Spectrometer (ICP–MS) facility is used for quantification of thirty two elements on a selected commercial bottled drinking water. A fully quantitative method was applied to check the concentration of thirty two elements (Li, Be, B, Na, Mg, Al, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Ag, Cd, Hg, Sb, Te, Ba, Tl, Pb, Bi and U) on the bottled water samples. Mercury was determined by using the flow injection mercury system (FIMS). The aim of the work is to check the quality of the bottled drinking water under investigation.

Key words: Bottled drinking water, ICP-MS, FIMS

INTRODUCTION

People can survive several days without food but only three to five days without water. The average adult consumes and excretes about 3 to 4 liters of water daily. The body uses water for digestion, absorption, circulation, transporting nutrients, building tissues, carrying away waste and maintaining body temperature.

During the last decade, the development of Inductively Coupled Plasma Mass Spectrometry (ICP–MS) made it one of the most important contributors to the inorganic analytical chemistry. It becomes a widely used and reliable technique for the analysis of water samples of different origin ^{1–8}.

Several metal ions such as sodium, potassium, magnesium, and calcium are essential to sustain biological life. At least six additional metals, chiefly transition metals, are also essential for optimal growth, development and reproduction; these are manganese, iron, cobalt, copper, zinc, and molybdenum, which are in small enough quantities to be considered trace elements. Trace metals function mostly as catalysts for enzymatic activity in human bodies. However, all essential trace metals become toxic when their concentration becomes excessive. Drinking water containing the above trace metals in very small quantities may actually reduce the possibility of deficiencies of trace elements in the diet. In addition to the metals essential for

^{*} Corresponding author. E-mail oaldayel@kacst.edu.sa

human life, water may contain toxic metals like mercury, lead, cadmium, chromium, silver, selenium, aluminum, arsenic, and barium. These metals can cause chronic or acute poisoning and should be eliminated from drinking water, if possible ^{9–13}.

Concerning the chemical quality of drinking water and health issues, most emphasis has been placed on excessive amount of various elements. It should be noted, however, that water related health problems can also occur due to element deficiencies. This problem is not covered by present day water regulations. Such deficiencies occur most often in rural communities in developing countries, where mainly locally produced water and food are consumed. Deficiency related health problems may result from low levels of elements such as F, I and Se in drinking water¹⁴.

The concentration of some inorganic trace metals is restrictive factor for drinking water quality, generally as consequence of their effects on the health. This fact makes necessary its regulation and monitoring ¹.

It is often assumed that natural water from deep wells is clean and healthy ¹³. This is usually true concerning bacteriological composition. The inorganic chemical quality of this water is, however, rarely adequately tested before the wells are put into production. Due to variations in the regional geology and water/rock interactions, high concentration of many chemical elements can occur in such water. During the last ten years, several studies have shown that wells in areas with particular geological features yield water that does not meet established drinking water norms without any influence from anthropogenic contamination ¹⁴.

Any natural material on earth pure water contains the whole periodic table of elements at least in traces. Natural concentrations in water from different parts of the world have recently been documented by Reimann and de Caritat¹⁵. Reported natural concentrations for many elements span several orders of magnitude. Presence and concentration of all elements, including the so called heavy metals, in all earth materials is a natural phenomenon and has in the first instance nothing to do with human interference, e.g. pollution. Water is a powerful solvent; we should thus not be surprised to find many elements in rather high concentrations in natural waters. However, even natural uncontaminated waters can contain trace constituents that are directly detrimental to health. This has been long documented for elements like F. More recently, very high values of As in ground water from India and Bangladesh, causing severe health problems on a large scale of people have been reported. Hungary is another country where high As in drinking water has been reported to cause severe health problems¹⁶.

The European Economic Community (Eu)¹⁰ and the World Health Organization (WHO)¹¹ recommended general guidelines for the quality of water used for human consumption. These guideline values were used, however, it is generally recognized that dietary intake is the main contributing factor to uptake of trace metals by man¹².

In the Gulf States, there is an overgrowth of the bottled drinking water business to cover the increasing demands of good drinking water quality. There is a concern of the quality of these bottle drinking water. In this work, the concentration of some toxic elements has been checked for twenty four bottled drinking water samples. Those are the most popular brand names in Riyadh area.

EXPERIMENTAL

Chemicals and reagents

High purity water (Specific resistivity 18 M Ω cm⁻¹) obtained from a Millipore Mili–Q water purification system was used throughout the work. A multi element standard (Merck–6) containing 30 elements with certified concentration values was used as an external standard during the analysis. The Standard Reference Material (SRM), Nist–1640 Natural Water purchased from the National Institute of Standards and Technology (NIST) USA.

Samples

Samples were selected as the most popular brand of bottled drinking water in Riyadh, Saudi Arabia. All bottles were made of Polyethylene or Polyethylene terephthalate. A full information about each samples is presented in Table 1.

Elements analysis

The analysis is performed by a Perkin–Elmer Sciex Instruments multi–element ICP–MS spectrometer, type ELAN6100, equipped with a standard torch, cross flow nebulizer and Ni sampler and skimmer cones. The typical instrument conditions and measurement parameters used throughout are listed below:

Value	Description
1.000	Nebulizer Gas Flow
8.250	Lens Voltage
1100.000	ICP RF Power
-2000.000	Analog Stage Voltage
1200.0	Pulse Stage Voltage
70.0	Discriminator Threshold
-6.500	AC Rod Offist

Hg analyses

A flow injection mercury system (FIMS) from Perkin Elmer FIMS-400 was used for determination of Hg in water samples.

Table 1. Samples information

S.	Name and	Produce and Origin	Bottle	Contents as mentioned on the bottle in ppm					
No.	Distributor			Ca	Mg	Na	K	Fe	PH
1.	Jeema, Al-Ahmari Est. Riyadh	Jeema, Mineral Water Dubai, UAE	**PET, 600 ml	4	2	32	2		7.2
2.	Najran, Najran Mineral Water Co.	Najran, Mineral Water Co. Najran, Saudi Arabia	Cup, *PE, 250 ml	18	5	28	1.5		7.2
3.	Hayat, Hayat Water Factory	Hayat, Water factory Alzulfi Saudi Arabia	Cup PE, 250 ml	10	3	25	1.3	0.01	7.1
4.	Mozn, ALjanob Water Factory	ALjanob, Water Factory sabya Saudi Arabia	PET, Bottle 300 ml	18	8	21	2.5	0	7.3
5.	Hony, Hony Health Water Factor	Hony, Health Water Factory Riyadh Saudi Arabia	PET, Bottle 330 ml	8.8	3.4	28.01	2	0.01	7.2
5.	Shallalat, Gulf Union Factory	Gulf Union Factory Riyadh, ALsseeli Saudi Arabia	PET, Bottle 600 ml	10	3	22	1.7	0	7.2
7.	Najran, Najran Mineral Water Co.	Najran, Minearl Water Co. Najran, Saudi Arabia	PET, Bottle 600 ml	18	5	28	1.5		7.2
3.	Evian, evian company	Evian, Company Evian France	PET, Bottle 500 ml	78	24	5	1		
).	Volvic, Volvic Company	Volvic, Compnay Volvic France	PET, Bottle 1500 ml	11.5	8	11.6	6.2		
10.	Hilwa, ALjouf Mineral water bottling company	ALjouf, Mineral Water bottling company Dawmat Aljandal Aljouf Saudi Arabia	PET, Bottle 600 ml	28.5	11.9	23.7	13.4	o II	7.4
1.	Music, Music Company	Music, Company Canada	PET Bottle, 400 ml	49.8	2.62	2.37	0.57	0.03	7.2
2.	Crystal	Crystal, Mineral Water Company Fujairah, UAE	PET, 1500 ml	10	5	20	1>		7.4
3.	ALqassim	ALqassim, Health Water Factory Alqassim ALbadaya Saudi Arabia	PET, Bottle, 650 ml	8.4	1	22.4	0.5		7.1
4.	Zulal	Zulal, Water Factory Riyadh Dammam Saudi Arabia	PET, Bottle 1500 ml	21.2	4.5	20	1.22	0.01	7.2
5.	Sohat	The societe des eaux minerals libanaises sal Falougha lebanon	PET, Bottle 500 ml	31.3	5.2	3.5	0.5	0.01	7.9
6.	Almanhal	Almanhal, Water Factory Co. Riyadh Saudi Arabia	PET, Bottle 500 ml	15	5	12	0.2	0.01	7
7.	Safa	Makkah, Water Company Makkah Saudi Arabia	PET, Bottle 500 ml	19	4	18	2		7.2
8.	Gulfa	Gulfa, Mineral Water Co. Ajman UAE	PET, Bottle 600 ml	4.8	6.7	14	1.1		7.7
9.	Alghadir	Alghadir, East for Food Industries Brood Alser Saudi Arabia	Bottle, 600 ml	16	2 12	18	2.3		
20.	Nova	HWB Co Plant in ALWasse's Nufou ALWassee Saudi Arabia	PET, Bottle 330 ml	10.6	5.71	17.11	1.47		7.28
1.	Alshifa	Alshifa, Health Water Factory ALHasa Saudi Arabia	PET, Bottle 500 ml	2.5	1	38	2		7.2
2.	Hana	National Plant for Healthy Water AlQassim Buraidah Saudi Arabia	PET, Bottle 600 ml	12	2.1	34	0.6		7.4
23.	Hada	Alhada, Water Co. Ltd. Makkah Saudi Arabia	PET, Bottle 630ml	13	4	20	0.8		7.15
24.	Seven Springs	Seven Spring, Inc USA	PET, Bottle 500 ml	13	1.6		0.53		7

The FIMS is a complicated technique depending upon synchronization of mechanical, chemical and optical operations. The system contains three major units namely the spectrophotometer coupled with the flow injection circuitry, the amalgamation unit and the computer unit for automated control of the operation and measurements. The FIAS program was optimized and the program is saved as "Mercury 2" in the computer.

Method name:	Mercury 2	Slit width:	0.7 nm
Technique:	FIAS-MHS	Read time:	15.0 s
Wavelength:	253.4 nm	Read Delay:	0.0 s
BOC time:	2.0 s	Signal type:	AA
Measurement:	Peak height	Calibration:	Linear, zero intercept

The FIMS program

Step	Time	Pump 1 speed	Pump 2 speed	Valve position	Read	Heat	Cool	Argon
Pre-fill	8	100	40	Fill			X	X
Step 1	5	100	40	Fill		X		X
Step 2	25	100	40	Fill			X	X
Step 3	20	0	40	Inject			X	X
Step 4	20	0	40	Inject			X	X
Step 5	10	0	40	Fill			X	X
Step 6	20	0	40	Fill	X	X		
Step 7	10	0	40	Fill			X	X
Step 8	1	0	0	Fill		10.00		

The blank contained 2% (v/v) H_2SO_4 , 2% (v/v) HNO_3 and approx. $1.0 \text{ mg L}^{-1} \text{ KMnO}_4$ in deionized water. All the measuring standard and sample solutions were stabilized in the same medium.

Quality assurance

To assess the analytical process and make a comparative analysis, Standard Reference Material (SRM), Nist–1640 Natural Water purchased from the National Institute of Standards and Technology (NIST) USA was analysed in the same manner as all water samples.

Table 2 compares the certified values and those obtained in this work by a semi-quantitative method. The results are generally in good agreement with the certified values.

Table 2. Reference material NIST-1640

Element	Th	is work	Co	Certified				
	Con. PPb	% RSD	Con. PPb	% RSD				
Li	49.8	1.50	50.7	2.76				
Ве	36.4	4.95	34.94	1.17				
В	264	8.60	301.1	2.03				
Na	27800	0.34	29530	1.05				
Mg	5800	0.50	5819	0.96				
Al .	47.4	0.32	52	2.88				
K	899	0.93	994	2.72				
Ca	6990	0.97	7045	1.26				
V	11.9	1.16	12.99	2.85				
Cr	37.1	0.45	38.6	4.15				
Mn	117	0.46	121.5	0.91				
Fe	35.2	15.65	34.3	4.66				
Co	19	0.50	20.28	1.53				
Ni	27	1.19	27.4	2.92				
Cu	89.2	0.19	85.2	1.41				
Zn	64.7	1.36	53.2	2.08				
Ga	0.198	30.20						
As	27.3	1.59	26.67	1.54				
Se	23.7	3.95	21.96	2.32				
Rb	2.22	1.65	2	1				
Sr	108	0.40	124.2	0.56				
Mo	43.5	2.04	46.75	0.56				
Ag	6.48	0.39	7.62	3.28				
Cd	21.9	2.14	22.79	4.21				
Sb	12.6	2.85	13.79	1.46				
Ге	0.328	26.89						
Ba	139	0.96	148	148				
ri	0.146	42.60	< 0.1*,					
Pb	26.8	0.79	27.89	0.50				
Bi	0.143	40.49						
U	0.834	7.41						

CONCLUSION

Table 3 shows the elemental concentration of thirty two elements on the samples under investigation. Non of the elements exceed the WHO guidelines limits for drinking water. Beryllium is found infrequently in drinking water and only at very low concentrations, usually less than 1 μg litre⁻¹. Beryllium appears to be poorly absorbed from the gastrointestinal tract. Beryllium and beryllium compounds have been classified by IARC (International Agency for Research on Cancer) as being probably carcinogenic to humans (Group 2A) on the basis of occupational exposure and inhalation studies in laboratory animals. There are no adequate studies by which to judge whether it is carcinogenic by oral exposure. The very low concentrations of beryllium normally found in drinking water seem unlikely to pose a hazard to consumers^{11, 16}.

Boron concentrations in water vary widely and depends on the surrounding geology and wastewater discharges. The guideline value of 0.5 mg litre⁻¹ is designated as provisional because, with the treatment technology available, it will be difficult to achieve in areas with high natural boron levels^{11, 16}. In this work, sample number 13 record the highest reading. It was 267 ppb but the WHO regulation for boron is 500 ppb.

Chromium is widely distributed in the earth's curst. It can exist in valences of +2 to +6. Total chromium concentrations in drinking water are usually less than 2.0 g litre⁻¹, although concentrations as high as 120.0 g litre⁻¹ have been reported. In general, food appears to be the major source of intake¹⁷. The maximum reading of Cr in the sample under investigation is 6.88 ppb.

The dominant source of nickel exposure in the non smoking is food, non occupationally exposed population; water is generally a minor contributor to the total daily oral intake. However, where there is heavy pollution or use of certain types of kettles, of non resistant material in wells, or of water that has stood for an extended time in water pipes, the nickel contribution from water may be significant.

As regards health risks, inhalation is an important route of exposure to nickel and its salts; IARC concluded that inhaled nickel compounds are carcinogenic to humans (Group 1) and metallic nickel is possibly carcinogenic (Group 2B). However, there is a lack of evidence of a carcinogenic risk from oral exposure to nickel 11, 16.

Copper concentrations in drinking water vary widely as a result of variations in pH, hardness, and copper availability in the distribution system. Levels of copper in running water tend to be low, whereas those of standing or partially flushed water samples are more variable and can be substantially higher, particularly in areas where the water is soft and corrosive. Adult intake of copper from food is usually 1–2 mg/day and may be considerably increased by consumption of standing or partially flushed water from a system that contains copper pipes or fittings.

Table 3. Elemental concentration in ppb

Sample	1	2	3	4	5	6	7	8	9
Li	10.3	2.65	7.05	1.15	16	6.1	2.62	8.23	10.7
Ве	<dl< td=""><td>0.145</td><td>0.0757</td><td>0.122</td><td>0.08</td><td>0.113</td><td><dl< td=""><td>0.14</td><td>0.166</td></dl<></td></dl<>	0.145	0.0757	0.122	0.08	0.113	<dl< td=""><td>0.14</td><td>0.166</td></dl<>	0.14	0.166
В	<dl< td=""><td><dl< td=""><td>109</td><td><dl< td=""><td>86</td><td>247</td><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td>109</td><td><dl< td=""><td>86</td><td>247</td><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	109	<dl< td=""><td>86</td><td>247</td><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	86	247	<dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>
Na	30400	14900	31000	23000	46900	55900	15800	7490	13000
Mg	104	6240	3210	8700	5490	1510	6720	30700	9390
Al .	18.4	38	31.4	24.3	20.3	10.6	6.81	3.56	5.52
K	1710	2750	1570	3030	2350	1000	2950	1250	6850
Ca	12400	37000	8580	17800	13500	5350	39400	87900	13000
V	0.332	2.57	0.823	9.25	0.844	0.526	2.84	0.219	4.74
Cr	1.77	0.681	6.88	3.02	4.18	2.86	1.78	1.86	1.36
Mn	0.596	<dl< td=""><td><dl< td=""><td>0.0268</td><td>5.62</td><td>3.71</td><td><dl< td=""><td>1.04</td><td>0.32</td></dl<></td></dl<></td></dl<>	<dl< td=""><td>0.0268</td><td>5.62</td><td>3.71</td><td><dl< td=""><td>1.04</td><td>0.32</td></dl<></td></dl<>	0.0268	5.62	3.71	<dl< td=""><td>1.04</td><td>0.32</td></dl<>	1.04	0.32
Fe	37.1	77.3	6.04	43.8	88.2	39.5	110	219	40.2
Co	0.0889	0.0222	0.0359	0.0534	0.285	0.0532	0.0632	0.0945	0.0201
Ni	0.17	0.0835	0.159	0.184	1.35	0.314	0.0403	0.0948	0.147
Cu	0.889	<dl< td=""><td>0.522</td><td>6.11</td><td>4.72</td><td>2.66</td><td>0.088</td><td>1.87</td><td>2.98</td></dl<>	0.522	6.11	4.72	2.66	0.088	1.87	2.98
Zn	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>
Ga	0.0158	0.0442	0.015	0.0099	0.152	0.00481	0.0337	0.0095	0.0158
As	0.131	0.92	0.12	0.48	0.171	0.156	0.84	0.357	4.16
Se	<dl< td=""><td>0.347</td><td>0.419</td><td>1.25</td><td>1.15</td><td>0.733</td><td>1.11</td><td><dl< td=""><td>0.244</td></dl<></td></dl<>	0.347	0.419	1.25	1.15	0.733	1.11	<dl< td=""><td>0.244</td></dl<>	0.244
Rb	0.856	0.466	0.972	0.289	1.2	0.285	0.444	0.203	6.33
Sr	101	206	139	199	193	77.5	214	375	57.6
Mo	0.98	2.21	0.565	1.9	0.609	0.251	1.92	0.778	0.481
Ag	0.243	0.0374	0.0128	1.07	0.0241	0.0129	0.0096	0.0067	<dl< td=""></dl<>
Cd	0.0802	0.0273	0.005	0.0193	0.0333	0.0163	0.0009	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>
Hg	<dl< td=""><td>ND</td><td>ND</td><td><dl< td=""><td><dl< td=""><td>0.0376</td><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	ND	ND	<dl< td=""><td><dl< td=""><td>0.0376</td><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td>0.0376</td><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	0.0376	<dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>
Sb	0.15	ND	ND	0.17	0.16	0.12	0.22	1.64	1.31
Те	<dl< td=""><td>0.011</td><td><dl< td=""><td><dl< td=""><td>0.0289</td><td>0.0729</td><td><dl< td=""><td>0.142</td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	0.011	<dl< td=""><td><dl< td=""><td>0.0289</td><td>0.0729</td><td><dl< td=""><td>0.142</td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td>0.0289</td><td>0.0729</td><td><dl< td=""><td>0.142</td><td><dl< td=""></dl<></td></dl<></td></dl<>	0.0289	0.0729	<dl< td=""><td>0.142</td><td><dl< td=""></dl<></td></dl<>	0.142	<dl< td=""></dl<>
Ва	1.57	13.1	3.5	2.47	3.09	1.02	13.4	113	0.205
Tl	0.0079	0.0046	1.83E-06	<dl< td=""><td><dl< td=""><td>2.83E-06</td><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td>2.83E-06</td><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	2.83E-06	<dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>
Pb	1.15	0.0547	0.129	0.593	0.656	0.0783	<dl< td=""><td><dl< td=""><td>0.034</td></dl<></td></dl<>	<dl< td=""><td>0.034</td></dl<>	0.034
Bi	0.0151	0.0112	0.0060	0.0052	0.0053	0.0031	0.0024	0.0015	0.0014
U	0.0158	1.13	0.0224	0.267	0.0636	0.0057	1.21	1.76	0.215

Table 3. Elemental concentration in ppb (continue)

Sample	10	11	12	13	14	15	16	17	18
Li	7.72	1.67	9.64	0.315	5.23	0.943	6.06	3.6	2.95
Ве	0.0619	0.171	0.205	0.0771	0.158	0.152	0.0947	0.0565	0.118
В	<dl< td=""><td><dl< td=""><td><dl< td=""><td>267</td><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>191</td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>267</td><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>191</td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td>267</td><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>191</td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	267	<dl< td=""><td><dl< td=""><td><dl< td=""><td>191</td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>191</td><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td>191</td><td><dl< td=""></dl<></td></dl<>	191	<dl< td=""></dl<>
Na	25800	9930	24000	32000	33200	3530	15800	19400	49700
Mg	13300	8500	3200	1620	5270	7190	4360	5380	2960
Al	11.1	106	8.76	9.11	15.1	8.8	17.9	13.1	9.98
K	13300	2470	833	1020	983	534	879	1970	1780
Ca	34600	20500	12500	11800	25800	38700	16300	24100	2410
V	0.58	0.622	0.664	0.917	0.696	0.0658	0.395	2.04	0.838
Cr	3	0.461	3.45	2.6	2.97	<dl< td=""><td>1.79</td><td>2.18</td><td>3.14</td></dl<>	1.79	2.18	3.14
Mn	1.82	0.0994	5.27	2.13	3.15	<dl< td=""><td>1.78</td><td>1.75</td><td>1.58</td></dl<>	1.78	1.75	1.58
Fe	132	64.2	73.3	68	97.9	107	76.2	104	31.8
Co	0.056	0.0449	0.0955	0.0377	0.0743	0.029	0.0333	0.0801	0.0182
Ni	0.104	0.131	0.6	0.412	1.91	<dl< td=""><td>0.449</td><td>2.16</td><td>0.0355</td></dl<>	0.449	2.16	0.0355
Cu	1.83	1.81	1.26	0.244	0.999	<dl< td=""><td>4.16</td><td>2.32</td><td>5.51</td></dl<>	4.16	2.32	5.51
Zn	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>23.5</td><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>23.5</td><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>23.5</td><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td>23.5</td><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>23.5</td><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td>23.5</td><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<>	23.5	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>
Ga	0.0116	0.0154	0.0066	0.0007	0.0109	0.0146	0.0085	0.0009	0.004
As	0.0761	1.29	0.137	0.0584	0.0712	0.0598	0.0603	0.0657	0.12
Se	1.01	0.311	0.887	1.01	0.468	0.104	0.631	0.96	0.15
Rb	15.9	0.293	0.948	0.29	0.659	0.294	0.446	0.156	0.534
Sr	267	57.3	198	175	216	48.6	162	118	31.7
Mo	2.42	1.14	0.171	0.234	0.238	0.576	0.325	1.84	0.0465
Ag	<dl< td=""><td>0.0133</td><td><dl< td=""><td>0.348</td><td>0.0193</td><td>0.0005</td><td>0.087</td><td>0.44</td><td>0.004</td></dl<></td></dl<>	0.0133	<dl< td=""><td>0.348</td><td>0.0193</td><td>0.0005</td><td>0.087</td><td>0.44</td><td>0.004</td></dl<>	0.348	0.0193	0.0005	0.087	0.44	0.004
Cd	0.0045	0.0149	0.003	0.0294	<dl< td=""><td><dl< td=""><td>0.0027</td><td>0.0023</td><td>0.0027</td></dl<></td></dl<>	<dl< td=""><td>0.0027</td><td>0.0023</td><td>0.0027</td></dl<>	0.0027	0.0023	0.0027
Hg	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>
Sb	0.58	0.35	0.16	0.16	0.14	1.07	0.12	0.16	0.11
Те	0.106	0.0497	0.0576	0.0375	<dl< td=""><td>0.0214</td><td>0.0261</td><td>0.0166</td><td>0.0452</td></dl<>	0.0214	0.0261	0.0166	0.0452
Ва	75.3	7.76	0.375	1.38	2.77	1.08	2.46	9.55	2.96
Tl	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>
Pb	0.0401	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>0.0546</td><td>1.16</td><td>0.224</td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>0.0546</td><td>1.16</td><td>0.224</td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td>0.0546</td><td>1.16</td><td>0.224</td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>0.0546</td><td>1.16</td><td>0.224</td></dl<></td></dl<>	<dl< td=""><td>0.0546</td><td>1.16</td><td>0.224</td></dl<>	0.0546	1.16	0.224
Bi	<dl< td=""><td><dl< td=""><td><dl< td=""><td>0.0012</td><td><dl< td=""><td><dl< td=""><td>0.001</td><td>0.0093</td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>0.0012</td><td><dl< td=""><td><dl< td=""><td>0.001</td><td>0.0093</td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td>0.0012</td><td><dl< td=""><td><dl< td=""><td>0.001</td><td>0.0093</td><td><dl< td=""></dl<></td></dl<></td></dl<></td></dl<>	0.0012	<dl< td=""><td><dl< td=""><td>0.001</td><td>0.0093</td><td><dl< td=""></dl<></td></dl<></td></dl<>	<dl< td=""><td>0.001</td><td>0.0093</td><td><dl< td=""></dl<></td></dl<>	0.001	0.0093	<dl< td=""></dl<>
U	0.0031	0.116	0.0025	0.209	0.024	0.275	0.0513	1.83	0.0017

Table 3. Elemental concentration in ppb (continue)

Sample	19	20	21	22	23	24	*WHO or **Eu	Max in Ref 14	Max in this work
Li	0.726	11.5	5.05	3.15	1.23	6.44		01.00	10.3
Ве	0.142	0.0394	0.113	0.0815	0.253	0.108		0.04	0.205
В	<dl< td=""><td>28.5</td><td>157</td><td>60.1</td><td><dl< td=""><td><dl< td=""><td>500</td><td></td><td>267</td></dl<></td></dl<></td></dl<>	28.5	157	60.1	<dl< td=""><td><dl< td=""><td>500</td><td></td><td>267</td></dl<></td></dl<>	<dl< td=""><td>500</td><td></td><td>267</td></dl<>	500		267
Na	26800	19600	48600	46400	26400	5920			55900
Mg	4010	5880	2130	5440	3880	2050		23178	30700
Al	5.04	4.15	19.2	7.67	5.11	10.8	200 Eu		106
K	1570	1330	2690	3010	1210	4400			13300
Ca	19500	16400	5310	17800	14500	15300			87900
V	1.48	0.994	1.05	0.631	2.57	3.46			9.25
Cr	3.77	1.07	4.69	2.47	2.08	6.43	50	3.8	6.88
Mn	<dl< td=""><td><dl< td=""><td>0.983</td><td>0.644</td><td>0.288</td><td>4.49</td><td>50</td><td>0.825</td><td>5.62</td></dl<></td></dl<>	<dl< td=""><td>0.983</td><td>0.644</td><td>0.288</td><td>4.49</td><td>50</td><td>0.825</td><td>5.62</td></dl<>	0.983	0.644	0.288	4.49	50	0.825	5.62
Fe	71.9	50.9	49.5	69.2	63.3	67.3	200 Eu	20.36	132
Co	0.0141	0.0297	0.0304	0.134	0.201	0.127			0.201
Ni	<dl< td=""><td>0.216</td><td>1.06</td><td>0.95</td><td>0.222</td><td>0.689</td><td>20</td><td>26.45</td><td>2.16</td></dl<>	0.216	1.06	0.95	0.222	0.689	20	26.45	2.16
Cu	0.216	0.473	1.89	6.09	0.117	3.35	2000	10.54	6.09
Zn	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>23.5</td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>23.5</td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>23.5</td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>23.5</td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td></td><td></td><td>23.5</td></dl<></td></dl<>	<dl< td=""><td></td><td></td><td>23.5</td></dl<>			23.5
Ga	0.0048	0.005	0.0086	0.006	0.0114	0.0211			0.044
As	0.0567	0.0676	0.128	0.0704	0.0572	2.71	10		4.16
Se	1.26	0.503	0.406	0.381	0.97	<dl< td=""><td>10</td><td>0.04</td><td>1.26</td></dl<>	10	0.04	1.26
Rb	0.597	1.19	1.64	3.19	0.145	2.91			15.9
Sr	237	194	78.2	174	48.2	51.3			375
Мо	0.752	0.163	0.196	0.095	0.364	0.355	70		2.41
Ag	0.0365	<dl< td=""><td>0.207</td><td><dl< td=""><td><dl< td=""><td>0.0007</td><td></td><td></td><td>0.44</td></dl<></td></dl<></td></dl<>	0.207	<dl< td=""><td><dl< td=""><td>0.0007</td><td></td><td></td><td>0.44</td></dl<></td></dl<>	<dl< td=""><td>0.0007</td><td></td><td></td><td>0.44</td></dl<>	0.0007			0.44
Cd	0.005	0.0045	<dl< td=""><td>0.0129</td><td><dl< td=""><td>0.0709</td><td>3</td><td>1.4</td><td>0.08</td></dl<></td></dl<>	0.0129	<dl< td=""><td>0.0709</td><td>3</td><td>1.4</td><td>0.08</td></dl<>	0.0709	3	1.4	0.08
Hg	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>1</td><td>2.58</td><td>0.0376</td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>1</td><td>2.58</td><td>0.0376</td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>1</td><td>2.58</td><td>0.0376</td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td>1</td><td>2.58</td><td>0.0376</td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>1</td><td>2.58</td><td>0.0376</td></dl<></td></dl<>	<dl< td=""><td>1</td><td>2.58</td><td>0.0376</td></dl<>	1	2.58	0.0376
Sb	0.15	0.23	0.13	0.12	0.20	0.52	5	1.06	1.64
Те	<dl< td=""><td><dl< td=""><td><dl< td=""><td>0.0533</td><td><dl< td=""><td>0.123</td><td></td><td></td><td>0.14</td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>0.0533</td><td><dl< td=""><td>0.123</td><td></td><td></td><td>0.14</td></dl<></td></dl<></td></dl<>	<dl< td=""><td>0.0533</td><td><dl< td=""><td>0.123</td><td></td><td></td><td>0.14</td></dl<></td></dl<>	0.0533	<dl< td=""><td>0.123</td><td></td><td></td><td>0.14</td></dl<>	0.123			0.14
Ва	9.57	1.33	0.845	6.97	4.27	29.2	700		75.3
Tl	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>0.007</td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>0.007</td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>0.007</td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>0.007</td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td></td><td></td><td>0.007</td></dl<></td></dl<>	<dl< td=""><td></td><td></td><td>0.007</td></dl<>			0.007
Pb	<dl< td=""><td><dl< td=""><td><dl< td=""><td>0.132</td><td><dl< td=""><td>0.0006</td><td>10</td><td></td><td>1.16</td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>0.132</td><td><dl< td=""><td>0.0006</td><td>10</td><td></td><td>1.16</td></dl<></td></dl<></td></dl<>	<dl< td=""><td>0.132</td><td><dl< td=""><td>0.0006</td><td>10</td><td></td><td>1.16</td></dl<></td></dl<>	0.132	<dl< td=""><td>0.0006</td><td>10</td><td></td><td>1.16</td></dl<>	0.0006	10		1.16
Bi	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>0.015</td></dl<></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>0.015</td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>0.015</td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td></td><td></td><td>0.015</td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td></td><td></td><td>0.015</td></dl<></td></dl<>	<dl< td=""><td></td><td></td><td>0.015</td></dl<>			0.015
U	1.51	0.0263	0.0903	0.651	0.001	0.0017	2		1.83

* WHO : World Health Organization EU : European Union directive guidelines

The IPCS (International Program on Chemical Safety) Task Group responsible for preparation of Environmental Health Criteria monograph for copper concluded that: "The upper limit of the AROI (Acceptable Range of Oral Intake) in adults is uncertain but it is most likely in the range of several but not many mg per day (several meaning more than 2 to 3 mg/day). This evaluation is based solely on studies of gastrointestinal effects of copper contaminated drinking water. A more specific value for the upper AROI could not be confirmed for any segment of the general population. The available data on toxicity in animals were considered not helpful in establishing the upper limit of the AROI, due to uncertainty about an appropriate model for humans" 11, 16, 18.

Arsenic is a naturally occurring element, ubiquitous in the environment in both; organic and inorganic forms. Inorganic arsenic, the more toxic form, is found in groundwater, surface water, and in many foods, such as fish, rice and grains. Human exposure to inorganic As is mainly through drinking water. According to a recent FDA Diet Study, the average adult As intake from food is 53 μ g/day, of which about 20% (13 μ g) is inorganic. (For perspective, at the currently approved MCL (Maximum Contaminated Level) of 50 μ g/L, total As intake from 2L water would be about 100 μ g, with the large majority of that being inorganic; at 10 μ g/L, total As would approximate 20 μ g, again mostly in the inorganic form)^{16, 19}. The highest concentration of arsenic found in this work is 4.16 ppb.

Selenium levels in drinking water vary greatly in different geographical areas but are usually much less than 0.01 mg/litre. Foodstuffs such as cereals, meat, and fish are the principal sources of selenium in the general population. Levels in food vary greatly according to geographical area of production.

The NOAEL (No Observed Adverse Effect Level) in humans was estimated to be about 4 μ g/Kg of body weight per day. The recommended daily intake of selenium is about 1 μ g/kg of body weight for adults^{11, 16}.

Molybdenum concentration in drinking water are usually less than 0.001 mg/litre. However, in areas near mining sites, molybdenum concentrations as high as 200 μ g/litre have been reported. Dietary intake of molybdenum concentrations is about 0.1 mg per day per person. Molybdenum is considered to be an essential element, with an estimated daily requirement of 0.1 – 0.3 mg for adults ^{11, 16}.

Cadmium is released to the environment in waste water, and diffuse pollution is caused by contamination from fertilizers and local air pollution. Contamination in drinking water may also be caused by impurities in the zinc of galvanized pipes and solders and some metal fittings, although levels in drinking water are usually less that $1 \mu g/litre$. Food is the

main source of daily exposure to cadmium. The daily oral intake is $10-35~\mu g$. Smoking is a significant additional source of cadmium exposure $^{11,~16,~18}$.

Antimony salts and possibly organic complexes of antimony are typically found in food and water at low levels. Reported concentrations of antimony in drinking water are usually less than 4 ppb. Estimated dietary intake for adults is about 0.02 mg/day¹¹.

Antimony concentration in polyethylene terephthalate (PET) is in the range of 136–190 ppm^{20, 21}. Most of the drinking water bottle is made of PET. In this work, antimony concentration if these water bottle has been checked, but it does not show contamination. The highest concentration of antimony was 0.11 ppb.

Mercury is present in the inorganic form in surface and ground waters at concentrations usually less than 0.5 μ g litre⁻¹. Levels in air are in the range of 2–10 ng m⁻³. Mean dietary intake of mercury in various countries ranges from 2 to 20 μ g per day per person. The kidney is the main target organ for inorganic mercury, whereas methylmercury affects mainly the central nervous system.

In 1972, JECFA (FAO/WHO) Joint Expert Committee on Food Additives established a provisional tolerable weekly intake (PTWI) of 5 μ g kg⁻¹ of body weight of total mercury, of which not more than 3.3 μ g kg⁻¹ of body weight should be present as methylmercury. In 1988, JECFA reassessed methylmercury, as new data had become available, and confirmed the previously recommended PTWI of 3.3 μ g kg⁻¹ of body weight for the general population, but noted that pregnant women and nursing mothers were likely to be at greater risk from the adverse effects of methylmercury^{11, 16}.

Lead is present in tapwater to some extent as a result of its dissolution from natural sources, but primarily from household plumbing systems containing lead in pipes, solder, fittings, or the service connections to homes.

Placental transfer of lead occurs in humans as early as the twelfth week of gestation and continues throughout development. Young children absorb 4–5 times as much lead as adults, and the biological half–time may be considerably longer in children than in adults. Lead is toxic to both; the central and peripheral nervous systems, inducing subencephalopathic neurological and behavioural effects. The IARC has classified lead and inorganic lead compounds in Group 2B (possible human carcinogen). According to the WHO guidelines for drinking water quality, lead should not exceed 10 ppb^{11, 16, 18}.

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