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ASSESSMENT OF PHYSICOCHEMICAL CHARACTERISTICS AND HEALTH RISK OF DRINKING WATER

Fazrul Razman Sulaiman^{a*}, Nur Fadhilah Mohd Rafi^a, Shytie Nur Shafiqah Kamarudin^a, Sharifah Norkhadijah Syed Ismail^b

^aFaculty of Applied Sciences, Universiti Teknologi MARA Cawangan Pahang, 26400 Bandar Tun Abdul Razak Jengka, Pahang, Malaysia

^bDepartment of Environmental and Occupational Health, Faculty of Medicine and Health Sciences, Universiti Putra Malaysia, 43450 UPM Serdang, Selangor, Malaysia

Graphical abstract

Abstract



This study investigates the physical and chemical properties of bottled water and tap water at a university campus in Pahang, Malaysia. A total of seven bottled water brands, consisting of natural mineral (NM) and packaged drinking (PD) types, were first randomly selected. Three source locations of tap water were also examined. All water samples were analysed for their physicochemical characteristics, including pH, electrical conductivity (EC), temperature (using a YSI multi-parameter), turbidity (using a turbidity meter) and selected trace metals, along with copper (Cu) and zinc (Zn) using graphite furnace atomic absorption spectroscopy (GFAAS). Results were then examined against World Health Organization (WHO) and Malaysian Ministry of Health (MMOH) guidelines for drinking water. Health risks associated with trace metal were estimated using the risk assessment model. Turbidity values for tap water (2.85-4.94 NTU) were slightly higher than bottled water (0.77-1.03 NTU). A low turbidity value (0.77-0.93 NTU) suggests the presence of effective water treatment processes for NM bottled water. A low concentration of EC (0.003-0.010 mS/cm) indicates demineralization of PD bottled water. Overall quality of the bottled water and tap water was in compliance with guidelines recommended by WHO and MMOH, posing a minimum health risk and remaining safe for consumption.

Keywords: Bottled water, tap water, water quality, trace metals, university population

Abstrak

Kajian ini dijalankan untuk menyiasat ciri-ciri fizikal dan kimia air botol dan air paip di sebuah kampus universiti di Pahang, Malaysia. Sejumlah tujuh jenama air botol terdiri daripada jenis mineral semulajadi (NM) dan minuman berpaked (PD) telah dipilih secara rawak. Tiga lokasi punca air paip turut diperiksa. Semua sampel telah dianalisis untuk ciri-ciri fizikokimia, termasuk pH, kekonduksian elektrik (EC), suhu (menggunakan YSI multi-parameter), kekeruhan (menggunakan meter kekeruhan) dan logam surih terpilih, kuprum (Cu) dan zink (Zn) menggunakan Spektroskopi Serapan Atom-Relau Grafit (GFAAS). Data yang diperoleh kemudian dibandingkan dengan garispanduan air minuman oleh Pertubuhan Kesihatan Sedunia (WHO) dan Kementerian Kesihatan Malaysia (KKM). Risiko kesihatan berkaitan logam surih telah dianggarkan menggunakan model penilaian risiko. Nilai kekeruhan untuk air paip (2.85-4.94 NTU) didapati lebih tinggi daripada air botol (0.77-1.03 NTU). Nilai kekeruhan yang rendah mencadangkan keberkesanan proses rawatan air untuk air botol. Kekonduksian elektrik (EC) yang rendah (0.003-0.010 mS/cm) menunjukkan penyahmineral air botol PD. Keseluruhan kualiti air botol dan air paip adalah mematuhi garispanduan yang dicadangkan oleh WHO dan KKM, mencadangkan risiko kesihatan yang minimum dan selamat untuk digunakan.

Kata kunci: Air botol, air paip, kualiti air, logam surih, populasi universiti © 2016 Penerbit UTM Press. All rights reserved

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*Corresponding author fazrul@pahang.uitm.edu.my

1.0 INTRODUCTION

The increasing development of human population density has increased demand for safe drinking water [1-2]. Consumers often complain about the taste of chemicals. These chemicals include chlorine, which is widely used to purify tap water [3]. Similarly, many consumers are concern about the aesthetics of the water, rather than the contents [4]. In particular, the content and potential health hazards from drinking water are also vital areas to be considered [5].

Bottled drinking water consumption has increased in the past three decades [3]. General interest in bottled drinking water began in the late 1970s and by the 1980s, retail sales of bottled water increased due to dynamic promotion campaigns [6]. Many consumers worldwide have turned to bottled water as their primary source of drinking water, including Malaysians [7]. Moreover, several cases of contamination of tap water have been reported in Malaysia [8]. This may have led to the increasing number of Malaysians consuming bottled water, although tap water is more reasonably priced.

In the Malaysian market, there are two types of bottled water, namely natural mineral (NM) and packaged drinking (PD) [7]. Natural mineral water bottles have blue or green coloured caps, while packaged drinking water is given a white cap. Natural mineral water originates from groundwater and emerges from a spring where it is tapped [9-10]. For packaged drinking water, a process such as reverse osmosis, distillation, or deionization is carried out to produce purified water [7].

The purpose of the study was to examine the physical and chemical characteristics of bottled water samples available at shops and tap water samples from selected location in a university campus in Pahang, Malaysia. The results were compared with drinking water quality guidelines set by the Malaysian Ministry of Health or MMOH [8] and the World Health Organization or WHO [11] to determine their suitability as drinking water. The health risks to human in associated with selected metals (copper and zinc) in the water samples were measured via risk assessment model.

2.0 MATERIALS AND METHODS

2.1 Sample Collection

A total of seven brands of commercial bottled water, consisting of natural mineral (NM) and packaged drinking (PD), were purchased randomly from two different retail stores between September and October 2013 with three replicates. All natural mineral (NM) bottled water originated from groundwater in Lengeng-Negeri Sembilan, Taiping-Perak and Karak-Pahang. The packaged drinking (PD) water was purified through distillation and reverse osmosis process, with water sources from Syabas-Selangor and Taiping-Perak. The brand names of the bottled water were kept anonymous, and code names were given to the samples throughout the study. All bottled water samples (NM and PD) were available in a 500 ml plastic bottle with plastic screw caps. Table 1 shows the classification of the bottled water.

As for comparison, tap water samples from three sampling points representing different important places in a university campus in Pahang were collected. The sampling sites were students' hostels, academic blocks, and laboratories (Figure 1b). The university campus is located in Jengka, Pahang, Malaysia, with a population of 10,000 for student and staff. Tap water samples were collected for two consecutive weeks in October 2013 with three replicates.

2.2 Sample Analyses

All the plastics and glassware were washed with 5% nitric acid (HNO₃) for at least 24 hours and rinsed thoroughly with ultrapure water type-1 (water sensitivity~18.2 Mohms · cm at 25°C). These steps were necessary to avoid any cross contamination. The water samples were divided into acidified and nonacidified subsamples. The non-acidified sample was used to analyse temperature, pH, and electrical conductivity (EC) using 556MPS YSI multi-parameter and turbidity using 2100P Turbiditimeter. All equipment was calibrated prior to use to ensure the accuracy of readings. For acidified samples, Whatman glass fibre filter paper (0.45 µm) was used to filter the water samples and then 5% HNO₃ was added to maintain pH 2. The samples were then preserved at 4°C before trace metal analysis. Copper (Cu) and zinc (Zn) were analysed using graphite furnace atomic absorption spectroscopy, GFAAS (Perkin Elmer PinAAcle 900T) with detection limits of 0.02 µg/l and 0.52 µg/l, respectively. External standard solutions were utilised as a reference for every sample. The regression coefficients (r^2) of the standard calibration curves for Cu and Zn were all above 0.99. Trace metal was analysed with triplicate to ensure precision in the results.



Figure 1 a) Sources of the analysed natural mineral bottled water b) Sampling locations for tap water samples in UiTM Pahang Malaysia

Table 1 Classification of bottled water samples

Sample	Туре	Number of samples	Water Resource	Remarks
S1	NM	3	Lenggeng-N. Sembilan	Normal treatment for groundwater
S2	NM	3	Taiping-Perak	Normal treatment for groundwater
S3	NM	3	Taiping-Perak	Normal treatment for groundwater
S4	NM	3	Karak-Pahang	Normal treatment for groundwater
S5	PD	3	Syabas-Selangor	Reverse osmosis
S6	PD	3	Taiping-Perak	Distillation
S7	PD	3	Taiping-Perak	Distillation

NM natural mineral, PD packaged drinking

2.3 Statistical Analyses

The results were analysed using IBM Statistical Package for Social Sciences (SPSS Version 21.0, USA). A one-way analysis of variance (ANOVA) procedure was applied to identify if there were any significant differences between natural mineral (NM) and packaged drinking (PD) bottled water. One-way ANOVA was used to examine any significant differences between three sampling locations of tap water. A Pearson's correlation analysis was utilised to determine any relationship between the analysed parameters. Principle component analysis (PCA) was carried out to determine the contribution of various factors of possible pollution sources as well as to interpret the relationships among variables.

2.4 Health Risk Assessment

Human may expose to chemical substances via three main pathways that are direct ingestion, inhalation and dermal absorption. In this study, the direct ingestion route i.e. oral intake and dermal route were considered for health risk assessment. Health risk assessment was performed through Hazard Quotient (HQ) calculation per Equation 1 [12]:

$$HQ = CDI/RfD \tag{1}$$

Hazard Quotient (HQ) is equivalent to exposure dose, expressed as chronic daily intake (CDI) in μ g/kg/day divided by reference dose (RfD) in μ g/kg/day. The exposure dose is separated into two; CDI ingestion is exposure dose contacted through ingestion of water and CDI dermal is exposure dose reached through dermal absorption. The chronic daily intake (CDI) was calculated using Equations 2 and 3 [12-15]:

 $CDI \text{ ingestion} = (CW \times IR \times ABSg \times EF \times ED) / (BW \times AT)$ (2)

 $CDI \text{ dermal} = (CW \times SA \times Kp \times ABSd \times ET \times EF \times ED \times CF) / (BW \times AT)$ (3)

where CW is average concentration of trace metal in water (μ g/I); IR is ingestion rate (2.2 I/day); SA is exposed skin area (2800 cm²); Kp is skin adherence

factor (cm/h); ABSd is dermal absorption factor; ABSg is gastrointestinal absorption factor; EF is exposure frequency (365 day/year); ED is exposure duration (70 years); ET is exposure time (0.6 h/day); CF is unit conversion factor for water (11/1000 cm³); BW is average body weight (70 kg); and AT is averaging time for non-carcinogens and carcinogens (25,550 days).

3.0 RESULTS AND DISCUSSION

3.1 Physicochemical Characteristics

Many factors influence the physical and chemical characteristics of natural water, such as atmospheric precipitation, residence time of the surface or groundwater, and mineralogy of the rock along the water path [16-17]. Table 2 shows the physicochemical characteristic of the water samples in this study. The pH values for natural mineral (NM) bottled water (between 7.64 and 7.95) were slightly lower compared to the packaged drinking (PD) bottled water samples (between 7.91 and 8.26). Nonetheless, the pH values for tap water were slightly higher than bottled water. There were significant differences in pH value between the NM and PD (p<0.05), but no significant differences were obtained between sampling locations for tap water (p>0.05), as seen in Table 3.

Dissolved carbon dioxide that forms carbonic acid in water determined the water pH [7]. The pH value does not have a direct effect on the consumer; however, it serves as an indicator of effective disinfection and water clarification [11]. The pH value should remain less than 8 for an effective disinfection. If the value is less than pH 6.5, there is the potential of trace metals such as Pb, Zn, and Cu released from the piping system [18]. This study has shown that the NM bottled water has an adequate disinfection system as the pH value remains below than 8. However, tap water and PD bottled water (S5 and S7) have values more than pH 8 (Table 2). Perhaps dissolution occurring in the water sources may affect the pH value, specifically hydrogen ion concentration [4].

The electrical conductivity (EC) values for bottled and tap water samples ranged between 0.001-0.130 mS/cm and 0.040-0.050 mS/cm, respectively. There were significant differences (p<0.05) for EC value between the NM and PD bottled water samples, and between sampling location for tap water samples. High EC value in the water sample indicates the presence of high dissolved solids content [19-20]. Samples of PD bottled water showed very low EC. The processes of reverse osmosis (RO) and distillation for the PD bottled water samples are effective in removing dissolved solids [7]. However, low EC value signifies extreme demineralization of PD bottled water and deficiency of minerals in the NM bottled water [7]. According to Mahajan *et al.* [21], long-term consumption of water with low mineral content may pose a number of health risks.

The average turbidity value for bottled water (0.77 to 1.03 NTU) was lower than tap water (2.85 to 4.94 NTU). Turbidity showed no significant differences (p>0.05) between the NM and PD bottled water. It has significant differences between sampling location for tap water samples (p<0.05). The turbidity value for tap water is higher compared to bottled drinking water possibly because of the treatment processes underwent by bottled water reduced the solid content in the water.

The average temperature for bottled water samples were range from 25.41 to 27.19 °C while for tap the range from 26.41 to 27.25 °C. Bottled water showed a larger range of temperature compared to tap water, probably because of the different source of water samples and different storage periods prior to consumption.

3.2 Trace Metals in Water Samples

The concentration of Cu and Zn in bottled water were between 1.50 - 11.23 μ g/l and 10.81 - 21.23 μ g/l respectively. However, the concentration of Cu (1.19-3.72 μ g/l) and Zn (14.53-19.60 μ g/l) in tap water samples were in lower range compared to the bottled water (Table 2). A one-way ANOVA analysis has shown significant differences of trace metals concentration between the bottled and tap water (p<0.05) (Table 3). There were also significant differences of trace metals concentration (p<0.05) between the NM and PD bottled water samples as shown in Table 3.

Cu in tap water possibly comes from brass fittings coated with chromium-nickel in the piping system [12]. The corrosion of the plumbing system contributes to the presence of trace metal such as Cu and Zn in tap water [2, 22]. Location of the water sources and the type of purification techniques imposed could result in high Cu and Zn concentration in bottled water [7]. The bottled water samples originally from groundwater in Taiping, Perak (S2 and S3) showed higher concentration of Cu. Syabas, Selangor had higher Zn concentration may cause toxic effects and produces an undesirable taste in the water and causes water to appear milky and cloudy [23]. Table 2 Water chemistry analyses for bottled water and tap water samples (mean ± SD)

	Parameter Parameter									
Sample	Туре	рН	EC (mS/cm)	Temperature (°C)	Turbidity (NTU)	Cυ (μg/l)	Zn (µg/l)			
S1	NM	7.95±0.04	0.13±0.00	27.19±0.26	0.93±0.11	9.51±0.04	16.99±0.10			
S2	NM	7.71±0.04	0.09±0.00	26.16±0.32	0.87±0.05	4.56±0.05	19.85±0.07			
\$3	NM	7.83±0.07	0.11±0.00	26.13±0.09	0.77±0.24	11.23±0.12	15.92±0.03			
S4	NM	7.64±0.05	0.13±0.00	25.41±0.16	0.93±0.20	2.82±0.04	11.96±0.03			
S5	PD	8.26±0.11	0.01±0.00	26.70±0.08	1.01±0.54	1.50±0.01	21.23±0.03			
S6	PD	7.91±0.14	0.001±0.00	26.04±0.11	0.87±0.07	1.63±0.03	16.32±0.05			
S7	PD	8.12±0.15	0.003±0.00	25.53±0.11	1.03±0.27	2.76±0.05	10.81±0.10			
SH	TW	8.11±0.05	0.04±0.01	26.41±0.54	2.85±0.25	1.19±0.01	14.53±0.55			
AB	TW	8.01±0.14	0.04±0.01	26.69±1.14	3.24±1.52	1.67±0.07	18.71±0.73			
Lab	TW	8.02±0.10	0.05±0.01	27.25±0.34	4.94±1.61	3.72±0.15	19.60±0.51			

AB academic block, EC electrical conductivity, NM natural mineral, PD packaged drinking, SD standard deviation, SH student's hostel, TW tap water

Table 3 Comparison of water chemistry with one-way ANOVA (p<0.05)

	p-value	p-value
Parameter	NM vs. PD	Three locations of tap water
рН	0.00	0.25
EC	0.00	0.03
Temperature	0.00	0.17
Turbidity	0.89	0.03
Cu	0.00	0.00
Zn	0.00	0.00

NM natural mineral, PD packaged drinking

3.3 Comparison with Water Quality Standard

The drinking water quality guidelines from the World Health Organization (WHO) [11] and the Malaysian Ministry of Health (MMOH) [8] were compared with physicochemical parameters of bottled water and tap water. Guler [24] stated that these guidelines are established for chemical constituents, microorganisms and physical characteristics that could pose a threat to human health. Table 4 shows the results for the measured parameters and the guidelines value. In general, all physicochemical parameters tested in this study were within the recommended values. These findings are in agreement with a previous study by Aris *et al.* [7]. Cu and Zn in the bottled and tap water were also well below the suggested limit.

3.4 Correlation Analysis and Principle Component Analysis (PCA)

Table 5 shows the Pearson's correlation between physicochemical characteristics of water samples and the metals (Cu and Zn) concentration. According to Taylor [25], the correlation coefficient, $r \leq 0.35$ represent weak correlations, r value of 0.36-0.67 indicate moderate links, and r value of 0.68-1.00 signify strong relationships. Bottled water samples show strong positive correlation between Cu and EC (r=0.68) and Zn has strong positive correlation with temperature (r=0.69). The pH value has a negative correlation with EC (r=0.67). For tap water samples, Cu showed a strong positive correlation with Zn (r=0.73) and moderate positive correlation with turbidity (r=0.63) as seen in Table 5. The strong correlations suggest

relationship between parameters, and this could propose a similar source of contribution factors.

Principal component analysis (PCA) was used to interpret the relationship between observed variables in the water samples. Mustapha [26] stated that PCA extracts eigenvalues and related factor loadings from the covariance matrix of the original variables produce new variables through varimax rotation. Factor loadings with value more than 0.75 categorized as 'strong', 0.50-0.75 as 'moderate', and less than 0.49 as 'weak' [26-27]. Thus, the important factors that explain the whole dataset could be identified [28-29].

Table 6 summarizes the rotational component matrix for both bottled and tap water samples. The PCA results for bottled water produced two main component factors that explained a total variance of 68.78% of the data. The component factors in which Factor-1 contributed 37.02% to the total variance with a high loading on EC (r = 0.920) and Cu (r = 0.760) (Table 6). EC and Cu perhaps are influenced by the similar geological contribution [17] although the samples came from different locations. Factor-2 contributed 31.76% to the total variance, with a high loading on temperature (r = 0.954) and Zn (r = 0.827). The temperature may be affected by the length of time of retention in the shop after production, indicating anthropogenic contribution. In specifies geologic input [30] though bottled water came from different sources. Hence, Factor-2 probably sees contributions from both man-made and geological factors.

In the case of tap water, the total cumulative variance for two factors was 78.19%. Factor-1 contributed 46.76% to the total variance with a high loading on Cu (r = 0.920) and Zn (r = 0.900). Cu and Zn

could result from the corrosion of plumbing systems [2, 22]. These trace metal could come from a similar source of lithogenic origin [1, 30]. The results indicate that Factor-1 may have both geologic and

anthropogenic contributions. Factor-2 contributed 31.423% to the total variance with a high loading on temperature (r = 0.860) suggesting the influence of anthropogenic activities.

Table 4 Comparison of physicochemical parameters of bottled water and tap water samples with WHO and MMOH guidelines for drinking water quality

Parameter		Bottled water		Tap water		
	Unit	Mean (NM)	Mean (PD)	Mean	WHO [11]	MMOH [8]
Cu	µg/l	7.03	1.96	2.19	2000	1000
Zn	µg/l	16.18	16.12	17.61	3000	3000
Turbidity	NTU	0.87	0.97	3.68	<5	NA
рН	-	7.78	8.09	8.04	6.5 – 8.5	6.5 – 9.0
EC	m\$/cm	0.12	0.002	0.04	NA	NA
Temperature	°C	26.22	26.09	26.78	NA	NA

NA no available standards, NM natural mineral, PD packaged drinking

Table 5 Correlation analyses of selected physicochemical characteristics of bottled water and tap water (strong correlation >0.68 is shown in bold)

Bottled water	рН	EC	Temp.	Turbidity	Cu	Zn
рН	1	-0.67**	0.34	0.28	-0.24	0.21
EC		1	0.12	-0.16	0.68**	-0.08
Temperature			1	0.08	0.39	0.69**
Turbidity				1	-0.22	-0.06
Cu					1	0.04
Zn						1
Tap water	рН	EC	Temp.	Turbidity	Cu	Zn
рН	1	-0.17	0.42	-0.51	-0.23	-0.46
EC		1	0.51	-0.14	0.54	0.49
Temperature			1	-0.24	0.47	0.28
Turbidity				1	0.63**	0.59
Cu					1	0.73**
Zn						1

** Correlation is significant at the 0.01 level (2-tailed)

Table 6 Factor loading for selected physicochemical characteristics of bottled water and tap water (strong PCA loading >0.750 is shown in bold)

	Bottled water	Bottled water	Tap water	Tap water
	Factor 1	Factor 2	Factor 1	Factor 2
рН	-0.768	0.393	-0.501	0.663
EC	0.920	0.056	0.621	0.539
Temperature	0.048	0.954	0.353	0.860
Turbidity	-0.436	-0.015	0.623	-0.631
Cu	0.760	0.392	0.920	0.129
Zn	-0.122	0.827	0.900	-0.035
Eigenvalues	2.221	1.906	2.806	1.885
Variance (%)	37.015	31.761	46.762	31.423
Cumulative (%)	37.015	68.776	46.762	78.185

3.5 Cu and Zn Health Risk Assessment

Human immunity system and cellular metabolism are related to Cu and Zn [21, 31-32]. Excessive intake of Cu and Zn can be harmful to human. For instance, excessive Zn intake in the body may lead to nausea, vomiting, epigastric pain, lethargy, and fatigue [33]. In contrast, deficiency of Zn may initiate an adverse effect on physical growth and neurodevelopment especially for young children [32]. Excessive intake of Cu may result in nausea, vomiting, and abdominal and muscle pain [34]. The HQ estimation was summarized in Table 7 based on the oral consumption of water and dermal absorption. Leung *et al.* [35] proposed that HQ>1 imply potential adverse health effects and HQ<1 signify little health effects. The HQ ingestion and HQ dermal of Cu and Zn were smaller than unity, suggesting that these metal elements may pose a minimum health effect to the university residents via oral consumption and dermal absorption. These results were in agreement with previous studies [12-13, 30].

The initial risk assessment engaged in this study comprises some possible reservations. The *RfD* obtained from USEPA might not be specific to Malaysia. Differences in exposure conditions and age might show different risks. Furthermore, the treatment processes underwent might change the effects of metal contaminants. This study only presents preliminary results of Cu and Zn, and an assessment of more trace metals (e.g. arsenic and cadmium) should be carried out to evaluate the risk precisely.

4.0 CONCLUSION

In this study, seven brands of bottled water and three locations of tap water in a university campus were assessed for the physical and chemical parameters. The pH value and turbidity for tap water were slightly higher than bottled water. However, the chemistry of bottled water may change during transportation or storage, particularly when containers exposed to sunlight or kept for an extended period of time. The bottled water samples that come from groundwater in Taiping-Perak were high with Cu concentration. Nevertheless, Cu and Zn concentrations in all water samples were below the limits recommended by WHO

and MMOH. Bottled water and tap water are perhaps influenced by geologic and anthropogenic contributions based on PCA results. Further analysis of health risk assessment found that Cu and Zn pose a minimum health hazard to the university population: therefore, they are safe to be consumed. Although the drinking water is regarded as safe, PD bottled water has extremely low values of EC indicating deficient in essential minerals due to the water treatment. This study presents baseline data for future reference especially for drinking water assessment. Analyses of mineral ions (e.g. calcium, sodium, potassium, magnesium), other trace metals (e.g. arsenic, cadmium and lead), and potentially carcinogenic substances are necessary for bottled water and tap water to maintain the safety of drinking water.

Table 7 Health risk assessment of Cu and Zn through oral ingestion and dermal absorption

Sample	Element	CDI ingestion (µg/kg/day)	CDI dermal (µg/kg/day)	RfD ingestion* (µg/kg/day)	RfD dermal* (µg/kg/day)	HQ ingestion	HQ dermal	∑HQ
NM	Cu	0.220	0.168	40	12	5.50 x10 ⁻³	1.40 x10 ⁻²	1.95 x10 ⁻²
	Zn	0.510	0.388	300	60	1.69 x10 ⁻³	6.50 x10 ⁻³	8.19 x10 ⁻³
PD	Cu	0.062	0.047	40	12	1.54 x10 ⁻³	3.91 x10 ⁻³	5.45 x10 ⁻³
	Zn	0.510	0.386	300	60	1.69 x10 ⁻³	6.50 x10-3	8.18 x10-3
TW	Cu	0.070	0.053	40	12	1.70 x10 ⁻³	4.41 x10-3	6.12 x10 ⁻³
	Zn	0.550	0.423	300	60	1.83 x10 ⁻³	7.00 x10-3	8.83 x10-3

NM natural mineral, PD packaged drinking, TW tap water, *USEPA [15]

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