Daily intakes of copper, zinc and arsenic in drinking water by population of Shanghai, China

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Abstract

Daily intakes of metals in drinking water are of extreme importance in risk assessment to human health. Some papers focused on this topic, but most of them did not consider the effect of age, gender and work location on daily intakes of metals in drinking water. The objective of present paper is to estimate the levels of Cu, Zn and As ingestion in drinking water in Shanghai, China and the effect of age, gender and work location on daily intakes of these metals. It was also the first time that such a detailed investigation on daily intakes of metals in drinking water was carried out in China in recent years. Drink/Sample (D/S) method was used to estimate the daily intakes of Cu, Zn and As in drinking water. The mean concentrations of Cu, Zn and As in drinking water were 10.8 µg/l, 0.29 mg/l and 0.91 µg/l, which were lower than U.S. EPA's Drinking Water Equivalent Level (DWEL) and WHO guideline values. The average daily intakes of Cu, Zn and As in drinking water was 21.12 µg/d, 0.65 mg/d, and 1.83 µg/d, which were on average 0.01%, 1.1% and 1.5% of the provisional maximum tolerable daily intake (PMTDI) set by the Joint FAO/WHO Expert Committee. There was a significant difference in daily intake of Zn for different work locations ($P=0.04$). But no other significant differences in daily intakes of metals in drinking water were found according to age, gender and work location. © 2005 Elsevier B.V. All rights reserved.

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1. Introduction

Human being's essential requisites of air, water and food contain trace amounts of a wide range of ele-

ments, some of which plays a significant role in biochemical processes (Pirrone and Keeler, 1996; Raghunath et al., 1997; Tripathi et al., 1999).

Because the levels of trace elements in food and water generally reflect trace metals burden in the environment, the amount of metals ingested through food and water is of extreme importance in risk assessment to human health (Barnard et al., 1997; Tripathi et al.,

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1997). It is also important to estimate the daily intakes of trace elements for populations in different geological locations in the world, where civilization level, living habit, economic situation and other factors are significantly different from each other. However, information on these topics is scarce, especially when dealing with developing countries.

The estimation of daily intakes of trace metals in drinking water should consider the concentration and dose ingested (Adgate et al., 2001; MacIntosh et al., 2001; Tsai et al., 2003). The estimation also depends on people's water consumption habits. Presently, most studies on daily intakes in drinking water rely on the water consumption hypothesis of Environmental Protection Agency (EPA) and World Health Organization (WHO) that may not correspond to the actual behavior of study population regarding tap water drinking. To accurately assess daily intakes in drinking water, it is important to obtain data on people's actual water consumption habits.

The objective of the present paper was to assess daily intakes of Cu, Zn and As in drinking water in a test population in Shanghai, China and the effects of gender, age and work location on daily intakes of those metals in drinking water. Another objective is to construct a database of water quality variables as well as water consumption habits for a representative sample of the population of Shanghai, China in the summer of 2003.

2. Materials and methods

2.1. Drink/sample method

Drink/Sample method (D/S method) created originally by studies on exposure to trace elements in food and beverages and later applied to drinking water consists of sampling each time an individual has a drink containing drinking water, and recording the volume he or she drank. If participants drank boiled tap water from the same thermos bottle, they should only take one sample. Due to time and money constraints, it was decided that only 163 volunteers would participate in this study. These volunteers were selected in a joint manner with Central Viewer Survey and Consulting Center-Taylor

Nelson Sofres (CVSC-TNS) Research Company. The volunteers were selected to represent blocks of residences, socio-economic levels in society and age groups in the population. Visits were carried out in July 2003 by qualified inquirers to evaluate an individual's past 24 h water consumption and explain how to fill in the questionnaire.

The water samples were stored in polyethylene bottles, pre-washed with concentrated nitric acid (1:1) and after collection, nitric acid (0.1% v/v) was added as a preservative (Chatterjee et al., 1993). The water was acidified with 50% v/v nitric acid.

2.2. Measurement methodology

The concentrations of Cu were determined by graphite furnace atomic absorption spectrometry (GFAAS, Perkin-Elmer 3100, Conn., USA). Concentrations of Zn were determined by flame atomic absorption spectrometry (Model Z-6100, Hitachi CO. Tokyo, Japan). Concentrations of As were determined by Atomic Fluorescence Spectrometer (AF-610A, BRAIC CO. Beijing, China). The concentrations of metals were determined according to the method described by QA/QC procedures employed by EPA for these types of operations and improved for the different stages mentioned (USEPA, 1991a,b, 1993, 1994, 1995, 1997).

Non-parametric methods were chosen for the data that did not follow a normal distribution using the Kolmogorov–Smirnov test. Mann–Whitney *U*-test was used in order to analyze differences between males and females for daily intakes. Mann–Whitney *U*-test was also used in order to analyze differences between different work locations. Kruskal–Wallis *H*-test was used in order to analyze differences among different age groups. A statistically significant difference was set at $P < 0.05$. Statistical analyses were conducted by means of SPSS for Windows (version 10.0).

3. Results and discussion

3.1. Volumes of drinking water

The volunteers were divided according to their age, gender and work location (Table 1). Water consump-

Table 1

Characteristics of volunteers and averages of drinking content and weight of volunteers in 24 h

	Number of volunteers	%	Daily water consumption (l/d)	Weight (kg)
AM'	81	49.7	2.1	66.9
AF	82	50.3	1.9	56.1
OM'	9	5.5	2.2	66.9
OF	9	5.5	2.0	61.0
MM'	55	33.7	2.1	67.8
MF	55	33.7	2.0	57.4
MM'-1	45	27.6	2.1	70.7
MF-1	37	22.7	1.9	55.7
MM'-2	10	6.1	2.1	63.4
MF-2	18	11.0	2.0	57.9
YM'	17	10.4	1.7	63.8
YF	18	11.0	1.7	50.0

AM': man; AF: woman; OM': old man; OF: old woman; MM': middle man; MF: middle woman; MM'-1: middle man who work inside the house; MF-1: middle woman who work inside the house; MM'-2: middle man who work outside the house; MF-2: middle woman who work outside the house; YM': young man; YF: young woman.

tion volumes according to age, gender and work location were also evaluated (Table 1). The mean consumption volume of drinking water was estimated to be 2.0 l/d, which was consistent with U.S. EPA's hypothesis (2 l/d). And it was higher than the general consumption of Canadians (1.6 l/d) and Americans (1.4 l/d) (Levallois et al., 1998). The volumes of water consumed by volunteers in different age, gender and work location groups were between 1.7 and 2.2 l/d.

3.2. Chemical results

Concentrations of Cu, Zn and As were listed in Table 2. Concentrations of Cu, Zn and As did not follow a normal distribution using the Kolmogorov–Smirnov test ($P < 0.05$). According to WHO, natural Cu concentrations in drinking-water are around a few micrograms per liter (WHO, 1981). Cu concentration in water distributed by copper pipes depends on such properties as hardness, pH, anion concentration, oxygen concentration, and temperature. (EPA, 1987; Slooff et al., 1989; Alam and Sadiq, 1989). The highest Cu concentration in tap water was reported to be 1.2 mg/l in Chile, 4.8 mg/l in USA, 0.1 mg/l in Saudi Arabia, and 1.9 µg/l in India. (Lagos et al., 1999; Al-SalehU and Al-Doush, 1998; Tarit et al., 2003). In our study, the highest Cu concentration

was 32.1 µg/l, which was much lower than U.S. EPA's Drinking Water Maximum Contaminant Level Goal (1.3 mg/l) (EPA, 2002; Kenneth and Gilbert, 2002) and WHO guideline value (2 mg/l) (WHO, 1993).

Zn concentration is usually low in drinking water, compared to the WHO guideline value (3 mg/l) (WHO, 1993). The Zn concentration in tap water can be much higher as a result of the leaching of Zn from piping and fittings (WHO, 1996). The mean Zn concentration in our study was 0.29 mg/l, which was lower than U.S. EPA's Drinking Water Equivalent Level (10 mg/l) and WHO guideline value for organoleptic properties (3 mg/l) (EPA, 2002; WHO, 1993). The highest Zn concentration was 1.63 mg/l, which was very close to the WHO guideline value (3 mg/l). The measured values for Zn in drinking water were compared to the results of other studies. Even higher Zn concentrations (up to 24 mg/l) were reported in a Finnish survey (WHO, 1996). A recent study showed that the mean Zn concentration in drinking water in West Bengal, India was 51.8 µg/l (Tarit et al., 2003).

According to WHO, inorganic As compounds are classified by IARC in Group 1 (carcinogenic to humans) on the basis of sufficient evidence for carcinogenicity in humans and limited evidence for carcinogenicity in animals (IARC, 1987; Luten et al., 1982). The level of As in natural waters generally varies between 1 and 2 µg/l (Hindmarsh and Mcurdy, 1986). In some parts of the world — Taiwan, western South America, Bangladesh, Inner Mongolia — As is present in high concentrations naturally in drinking water, at levels ranging from several hundred to well over 1000 µg/l. In some parts of the United States, the population is also exposed to levels of As in drinking water higher than 50 µg/l (Kenneth and Gilbert, 2002). The mean As concentration in the water samples in Shanghai was much lower than that in those countries and ranged from 0.00 to 5.98 µg/l, with a mean concentration of 0.91 µg/l and a standard devi-

Table 2

Water quality variables measured in drinking water samples from Shanghai, China, 2003

Parameters	Mean	S.D.	Maximum	Minimum	n
Total Cu (µg/l)	10.8	0.4	32.1	1.3	195
Total Zn (mg/l)	0.29	0.04	3.00	ND	188
Total As (µg/l)	0.91	0.05	6.00	ND	189

ation of 0.05. The current WHO recommended Guideline Value for As in drinking water is 10 µg/l, whereas many developing countries (including China) still have a previous value used by WHO, of 50 µg/l. The standard value for arsenic in drinking water in Australia has been set at 7 µg/l (NHMRC, 1996). It was important to note that the maximum As concentration in our study was very close to Australia's standard.

3.3. Daily intakes of Cu, Zn and As in drinking water

Table 3 showed the results of daily intakes of Cu, Zn and As in drinking water. Cu is ingested by humans mainly through food and drinking water. Drinking-water can contribute a significant proportion of the daily Cu intake if it has flowed through copper installations (EPA, 1987; Slooff et al., 1989). A provisional maximum tolerable daily intake (PMTDI) of 0.5 mg/d/kg of body weight was established by Joint FAO/WHO Expert Committee on Food Additives (JECFA) (WHO, 1982a,b). According to WHO, an allocation of 10% of the PMTDI to drinking-water gives a guideline value of 50 µg/d/kg. In our study, the average daily intake of Cu in drinking water was 21.12 µg/d which was only 0.01% of PMTDI for

drinking water and food. The recommended daily intake of copper in the United Kingdom is 1.2 mg for adults. The recommended dietary intake in the United States is 1.5–3.0 mg for adults (FAB, 1989; WHO, 1996). The average daily intake of Cu contained in drinking water by Shanghai sample population was 1.8% of the United Kingdom recommendations for minimum total ingestion of Cu for adults. In west Bangladesh, India the daily intake of Cu in drinking water was about 0.92 µg/d, which was much less than that in our study (Tarit et al., 2003).

Under certain circumstances, tap water can provide up to 10% of the daily intake of Zn (Gillies and Paulin, 1982). JECFA proposed a daily dietary requirement of Zn of 0.3 mg/kg of body weight and a provisional maximum tolerable daily intake (PMTDI) of 1.0 mg/kg of body weight (WHO, 1982a,b). WHO recommended that the daily requirement of Zn for adult humans is 15–22 mg/d (WHO, 1996). In our study, the daily intake of Zn was 0.66 mg/d, which was 1.1% of the PMTDI. It was only 4.4% of the WHO recommendations for minimum total ingestion of Zn for adults. It was important to note that the maximum daily intake of Zn through drinking water was 10.23 mg/d in our study, which was 68.2% of the WHO recommendations for minimum total ingestion of Zn for adults. A recent study showed that the daily intake of Zn in drinking water was about 0.16 mg/d in west Bangladesh, India (Tarit et al., 2003).

A critical source of As intake for the general population is the drinking water. A value of 13 µg/l may be derived (assuming a 20% allocation to drinking-water) on the basis of the provisional maximum tolerable daily intake (PMTDI) of inorganic As of 2 µg/kg of body weight set by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) in 1983 (WHO, 1989). In our study, the average daily intake of As was 1.83 µg/day, which was 1.5% of the PMTDI. Results from west Bangladesh survey showed that in arsenic affected area, water contributes about 75% of daily intake of As (Tarit et al., 2003).

Our results suggested that ingestion of these metals in drinking water were associated with a non-significant potential threat to human health for the general population of Shanghai, China.

Table 3 also showed the results of daily intakes of Cu, Zn and As in drinking water in segments of the

Table 3
Daily intakes of Cu, Zn and As in drinking water by general population of Shanghai

	As (µg/day)	Cu (µg/day)	Zn (mg/day)
All	0.65 ± 1.27	21.12 ± 12.63	1.83 ± 1.36
AM'	1.88 ± 1.34	21.32 ± 12.55	0.69 ± 1.42
AF	1.77 ± 1.39	21.01 ± 12.79	0.62 ± 1.12
OM'	2.27 ± 1.34	24.47 ± 10.32	0.72 ± 0.83
OF	2.85 ± 2.43	24.44 ± 15.44	0.53 ± 0.27
MM'	1.95 ± 1.46	21.98 ± 13.80	0.77 ± 1.66
MF	1.70 ± 1.28	20.11 ± 18.43	0.70 ± 0.99
MM'-1	2.07 ± 1.58	22.69 ± 15.92	0.82 ± 2.38
MF-1	1.92 ± 1.43	20.85 ± 11.44	0.65 ± 0.95
MM'-2	1.41 ± 1.23	18.79 ± 13.14	0.71 ± 0.58
MF-2	0.94 ± 0.86	18.58 ± 6.44	0.81 ± 1.63
YM'	1.45 ± 0.67	23.47 ± 18.43	0.42 ± 0.55
YF	1.46 ± 0.80	16.04 ± 7.39	0.42 ± 0.99

AM': man; AF: woman; OM': old man; OF: old woman; MM': middle man; MF: middle woman; MM'-1: middle man who work inside the house; MF-1: middle woman who work inside the house; MM'-2: middle man who work outside the house; MF-2: middle woman who work outside the house; YM': young man; YF: young woman.

population. It was important to note that gender, age and work location affected the daily intakes of Cu, Zn and As in drinking water. Our results showed that older people had significant higher daily intakes than the younger ones. But no significant differences in daily intakes of Cu, Zn and As among different age groups were found ($P > 0.05$). For example, the daily intake of As was 2.66 $\mu\text{g/d}$ for over 64 years, versus 1.85 $\mu\text{g/d}$ for 20–64 years old and 1.46 $\mu\text{g/d}$ for less than 20 years. This was largely due to the fact that older people drink more water than younger ones. Data on daily intakes according to the gender of the consumer was also assessed. In the present study we also could not find gender related differences ($P > 0.05$). But there was a tendency for males to have higher mean daily intakes of Cu, Zn and As than females (21.32 $\mu\text{g/d}$ for Cu, 0.69 mg/d for Zn and 1.88 $\mu\text{g/d}$ for As vs. 21.01 $\mu\text{g/d}$ for Cu, 0.62 mg/d for Zn and 1.77 $\mu\text{g/d}$ for As). Our results indicated that the difference of water consumption between males and females could be considered as an important factor. In general, males consumed more water than females (2.1 l/d vs. 1.9 l/d). Work location also affected the daily intakes of metals in drinking water. For example, middle aged people who worked inside the house had lower daily intakes of Zn than those who worked outside the house (0.74 mg/d vs. 0.76 mg/d). There was a significant difference in daily intake of Zn for different work locations ($P = 0.04$). But No such difference was found for daily intakes of Cu and As ($P > 0.05$). The non-significant results in our study may be due to our small samples.

4. Conclusion

Daily intakes of Cu, Zn and As in drinking water by Shanghai people were on average 0.01%, 1.1% and 1.5% of the provisional maximum tolerable daily intake (PMTDI) set by the Joint FAO/WHO Expert Committee. That is to say that ingested metals in drinking water contributed to a minor fraction of the total metals intake, and current drinking water levels of metals were associated with a non-significant potential threat to human health for general population of Shanghai, China. Our results also showed that gender, age and work locations all affected drinking water consumption as well as daily intakes of metals

in drinking water, but there were no statistically significant differences.

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