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Research Article

The Total Arsenic Concentrations of Aquatic Products and the Assessment of Arsenic Intake from Aquatic Products in Guangzhou, China

[1,2]Yu Guang-Hui, [2,3]Zhang Lei, [2]He Shu-You, [2]Wen Yan-Mao, [1]Zhang Yong and [1]Zhu Jia-Wen
[1]The School of Architecture and Urban Planning, Hunan Science and Technology University, Xiangtan 411201, P.R. China
[2]The School of Environmental Science and Engineering, Sun Yat-Sen University, Guangzhou 510275, P.R. China
[3]Department of Environmental Science and Engineering, ZhongKai University of Agriculture and Technology, Guangzhou 510225, P.R. China

Abstract: The aim of this study was to assess the contribution of aquatic products consumed by the resident to the daily dietary arsenic intakes of the residents of Guangzhou of Guangdong province in China. All aquatic products were sampled from supermarkets and terminal markets. Accuracy was assured using standard reference material (GBW08551) and recovery experiments. Total arsenic concentrations of aquatic products were determined after acid digestion by hydride generation atomic fluorescent spectrometry. A wide range of arsenic concentration (0.0075-1.2017 mg/kg) was found among the various aquatic products, the mean arsenic concentration in aquatic production was 0.2022 mg/kg. The arsenic concentrations of various aquatic products groups were as follows: Crustacean (0.3176±0.2324 mg/kg) >Mollusk fish (0.1979±0.2013 mg/k) >Saltwater fish (0.1558±0.1119 mg/kg) >Freshwater fish (0.1374±0.0970 mg/kg). The range of daily dietary arsenic intake of various residents through the consumption of aquatic products was 5.96-11.85 μg/day. The freshwater fish had the largest contribution to the daily dietary arsenic intakes from aquatic products in all type aquatic products, accounted for around 50%.

Keywords: Aquatic production, arsenic concentration, arsenic intake, Guangzhou

INTRODUCTION

Arsenic was a toxic element, but it was widely distributed in the environment around people. Contamination of heavy metals represented one of the most pressing threats to environmental resources and human health. World Health Organization (WHO) and the United States Environmental Protection Agency (EPA) thought Arsenic (As) as a known carcinogen. Arsenic has been classified as a carcinogen to humans by the International Agency for Research on Cancer (IARC) (Tapio and Grosche, 2006; Tsuda *et al*., 1992). While WHO and the Environmental Protection Agency (EPA) regulated water sources of arsenic, lack of strict regulations on food, beverages and air quality can lead to increased arsenic exposure (Wilson *et al*., 2012). The main cause of arsenic pollution in the environment was the high concentration of arsenic in soil parent material and the use of industrial manufacturing, mining and agricultural pesticides, disinfectants, fungicides and herbicides (Diaz *et al*., 2004; Kar *et al*., 2013). The soil, drinking water and food were subjected to serious pollution of heavy metal and arsenic in China, the arsenic pollution of food and the risk to human health have been more and more attention (Rötting *et al*., 2014; Wang *et al*., 2012; Christina and Andrea, 2014). Research showed that China was one of the countries with serious arsenic pollution. In Xinjiang, Inner Mongolia, Hunan, Guangdong province and some place had been the emergence of endemic arsenic poisoning. Xiao *et al*. (2009) found that the arsenic concentration in major grain and oil crop exceeded 32.2 and 34.8% than the limit of arsenic in food of China in arsenic polluted area. The current study paid more attention to the regional environmental arsenic pollution and arsenic concentration of soil, also the uptake, translocation and biomagnifications of arsenic in several food crops and vegetables. But combined with the research on human health was relatively less. The arsenic intake of resident depended on the arsenic concentration of food ate by resident and the dietary structure.

This research studied total arsenic concentration of market aquatic products and the daily intakes arsenic from aquatic products of various residents in Guangzhou and assessed the safety of arsenic concentration of food and the daily intakes arsenic from aquatic products. The results provide scientific basis for

Corresponding Author: Yu Guang-Hui, The School of Architecture and Urban Planning, Hunan Science and Technology University, Xiangtan 411201, P.R. China

Table 1: Percent recovery of total arsenic of vicious sample

		Percent of recovery (%)	
Sample	N	Range	Mean±S.D.
GBWZ19001-94 (bovine liver)	5	91.4-99.1	94.3±3.6
Ctenpharyngodon idellus	5	92.3-97.7	95.7±2.4
Penaeus orientalis	5	97.8-103.2	98.4±4.7
Loligo chinensis	5	92.3-98.1	95.7±3.0

health risk assessment and reasonable dietary of various residents in Guangzhou.

MATERIALS AND METHODS

Samples preparation: Sampling sites located in the wholesale markets, farmer's markets and super markets which sell most of the vegetables, aquatic products and fruits in Guangzhou, including 3 wholesale market, 6 farmer's markets and 4 supermarkets of aquatic products. The farmers market located the different administrative region of Guangzhou. The supermarket is a relatively large market food sale. Random samples were collected in each sampling site. Acquisition with a sample 3-5 at each sampling location and then mixed into a sample. Samples were frozen immediately on laboratory refrigerator after being collected.

Samples were washed 3-5 times with tap water. Then samples were rinsed 2-3 times with distilled water. Then samples were rinsed 3 times by ultra pure water. The samples were dried by the clean dry gauze or coarse filter paper. The edible parts of dried samples were taken down and chopped by stainless steel knife. At last samples were crushed into homogenate by food disposer.

Total arsenic analysis:

Method: The samples were digested in nitric acid and perchloric acid mixture. Samples were analyzed by atomic fluorescence spectrometer (GB/T5009.11-2003, 2003).

Measuring instrument: Double channel atomic fluorescence spectrophotometer AFS-610 (Beijing Titan Instruments Co. Ltd.).

Analysis conditions: (1.5% KBH_4) (0.5% NaOH). Valence: tetravalent. PH: 10% (V/V) HCl. Carrier gas: Ar (99.99%). Traffic: 0.5 L/min, Shielding gas: Ar (99.99%). Traffic: 1 L/min. Lamp current: 70 mA. Temperature: 200°C. The observation height: 8 mm. Detection limit: <0.06 μg/L. Precision: <2%. The linear range: 0.2 to 200 μg/L.

Quality control: All glassware was treated with Pierce solution (20% v/v).

Throughout the experiment, the quality assurance and quality control of measurement were checked by analyzing the standard reference material (provided by National Standard Substances Center, GBWZ19001-94, bovine liver) with each batch of sample. Duplicate analyses were performed for each sample. The standard addition recovery experiments were done for testing analysis method. The percent recovery of total arsenic of vicious aquatic products was showed as for Table 1.

Dietary survey: Dietary survey methods of this study included the accounting method, survey method and weighing method. This study investigated 450 university students (13-31 years) of 5 colleges and universities, 541 children (3-6 years) of 10 kindergartens, 427 old peoples (62-70 years) of 8 nursing homes and 601 workers (18-51 years) of 5 factories and 2 building sites. Investigation of dietary of standard man and different income groups used the data of the nutritional status of residents in Guangdong province in 2002. The standard man was adult male engaged in light physical activity. Low income group referred to the people who hers average annual family income was less than 2000 Yuan. Middle income group referred to the people who hers average annual family income was 2000-10000 Yuan. High income group referred to the people who hers average annual family income was more than 10000 Yuan (Ma *et al*., 2005).

Statistical analysis: The average value and standard difference represents the concentration of arsenic in aquatic products. The average concentration of arsenic refers to the average values of all the samples. Data analysis and statistics were performed by using Excel and SPSS 12.0.

RESULTS AND DISCUSSION

Total arsenic concentration in aquatic products: In Table 2 the number of samples, range, means, standard deviations and variation coefficient were presented. As shown in Table 2, there were great differences of arsenic concentration in various types of aquatic products. For freshwater fish, the arsenic concentration of all samples ranged between 0.0090-0.7453 mg/kg. Arsenic concentration was highest for *Siniperca chuatsi*, 0.2795 mg/kg. Arsenic concentration of *Clarias batrachus* was lowest, 0.0627 mg/kg. The variation coefficient range of all samples was 22.5-141.0%. The variation coefficient of arsenic concentration of *Monopterus albus* was maximum, 141%. The *Pangasius hypophthalmus* had the smallest variation coefficient, 22.5%.

For sea fish, the range of arsenic concentration was 0.0075-0.9654 mg/kg and the arsenic concentration of sea fish had no statistically significant difference. The highest concentration was *Hairtail*, 0.2417 mg/kg and the lowest concentration was sardine, 0.1085 mg/kg. The range of variation coefficient of all samples was 39.9-112.0%. The minimum variation coefficient was *Cololahis saira*, the maximum variation coefficient for *Sillago sihama.*

Table 2: Total arsenic concentration in vicious aquatic products

Aquatic products	n	Range	Mean	S.D.	C.V. (%)
Freshwater fish					
Cyprinus carpio	8	0.0124-0.2420	0.0743b	0.0401	54.0
Carassius auratus	6	0.0214-0.3035	0.1656ab	0.1051	63.5
Ctenpharyngodon idellus	8	0.0171-0.2974	0.1413ab	0.1374	97.2
Cyprinus pellegrir	9	0.0090-0.1902	0.0874b	0.0717	82.0
Cirrhina molitorella	6	0.0396-0.1620	0.0674b	0.0318	47.2
Cobitis taenia	6	0.0120-0.3036	0.1718ab	0.0941	54.8
Monopterus albus	7	0.0210-0.4847	0.1614ab	0.2279	141.0
Tilapia mossambica	8	0.0091-0.2756	0.1498ab	0.0891	59.5
Lateolabrax japoincus	7	0.0157-0.1965	0.1475ab	0.0712	48.3
Siniperca chuatsi	5	0.0312-0.7453	0.2795a	0.2325	83.2
Clarias batrachus	6	0.0178-0.1954	0.0627b	0.0314	50.1
Pangasius hypophthalmus	6	0.0357-0.1742	0.1397ab	0.0314	22.5
Total	82	0.0090-0.7453	0.1374B	0.0970	67.0
Saltwater fish					
Decapterus maruadsi	7	0.0147-0.2567	0.1478a	0.1142	77.3
Cololahis Saira	6	0.0246-0.1874	0.1641a	0.0654	39.9
Thunnus albacares	5	0.0750-0.1721	0.1257a	0.1187	94.4
Cantherinus modestus	6	0.0162-0.2874	0.1532a	0.0714	46.6
Trichiurus haumela	8	0.0214-0.9654	0.2417a	0.2118	87.6
Pseudosciaenu polyactis	8	0.0075-0.2341	0.1274a	0.0796	62.5
Taius tumifrons	6	0.0198-0.4874	0.1463a	0.1295	88.5
Sillago sihama	8	0.0231-0.3657	0.1085a	0.1214	112.0
Harpodon nehereus	5	0.0275-0.2257	0.1542a	0.0821	53.2
Nemipterus virgatus	5	0.0084-0.4687	0.1889a	0.1247	66.0
Total	64	0.0075-0.9654	0.1558B	0.1119	73.0
Crustacean					
Eriocheia sinensis	6	0.0219-0.7035	0.2277b	0.2336	103.0
Portunus pelagicus	5	0.1341-0.8136	0.1751c	0.1324	75.6
Anodonta woodiana	5	0.1471-0.7751	0.3821ab	0.2768	72.4
Palaemon carinicauda	8	0.0214-0.6263	0.2687b	0.2359	87.8
Penaeus orientalis	6	0.0274-0.3632	0.2842b	0.1496	52.6
Meretrix meretrix	7	0.2096-1.2017	0.5675a	0.3662	64.5
Total	37	0.0219-1.2017	0.3176A	0.2324	76.0
Mollusk fish					
Rhopilema esculenta	7	0.0296-0.2784	0.1456b	0.1268	87.1
Loligo chinensis	5	0.0124-0.3995	0.1562b	0.1421	91.0
Octopus vulgaris	6	0.0278-0.4842	0.2419a	0.2321	95.9
Sepiella maindroni	7	0.0095-0.7241	0.2477a	0.3041	123.0
Total	25	0.0095-0.4842	0.1979B	0.2013	99.0
Average		0.0075-1.2017	0.2022	0.0810	40.1

Different letters in each line meant significant difference at 5% level; S.D.: Standard deviation; C.V.: Coefficient of variation

Table 3: The daily intakes arsenic from aquatic products of various residents in Guangzhou (unit: μg/day)

Food	Standard man	Low income group	Middle income group	High income group	Elder	Child	University man	Worker
Freshwater fish	5.84	3.43	4.20	6.13	5.29	2.93	4.65	3.56
Seawater fish	2.21	1.30	1.59	2.32	2.00	1.11	1.76	1.35
Crustacean	2.70	1.59	1.94	2.84	2.45	1.36	2.15	1.65
Mollusk fish	1.12	0.66	0.81	1.18	1.02	0.56	0.89	0.68
Total	11.86	6.97	8.53	12.50	10.80	5.96	9.45	7.24

The arsenic concentration of crustacean ranges between 0214-1.2017 mg/kg. There were statistically significant differences of arsenic concentration between various crustaceans. The highest arsenic concentration was *Meretrix meretrix*, for 0.5675 mg/kg, significantly higher than that of other crust fish. The arsenic concentration of *Portunus pelagicus* was the lowest, for 0.1751 mg/kg. The range of variation coefficient was 52.6-103.0%. The minimum variation coefficient was *Penaeus orientalis*, the maximum variation coefficient for *Eriocheia sinensis*.

For mollusk fish, the range of arsenic concentration was 0.0095-0.7241 mg/kg. There were obvious differences between the various water products. Four kinds of mollusk fish had a larger range of variation coefficient (87.1-123.0%). The maximum variation coefficient was *Sepiella maindroni* and the minimum is *Rhopilema esculenta*.

In all samples, *Siniperca chuatsi* had one samples (0.7453 mg/kg) exceeded the limit of the arsenic concentration of food (GB4810-94, 1994) and exceeded 49% of the standard. *Monopterus albus* had one sample (0.4847 mg/kg) closed to the limit of arsenic in food.

There had not the limit of the total arsenic concentration of food about saltwater fish, crustacean and mollusk fish in China. The limit of the arsenic concentration of saltwater fish, crustacean and mollusk fish was inorganic arsenic in China and the limits value was 0.5 mg/kg. The new limit of the arsenic concentration of food also was in organic arsenic concentration to all aquatic products (GB/2762-2012, 2012).

In all, the statistically significant difference was observed in various types of aquatic products. Crustacean had the highest concentration of arsenic and freshwater fish has the lowest concentration of arsenic in all aquatic products. With the exception of a few samples, all aquatic products were safety to human health of residents in Guangzhou.

Intake arsenic from aquatic products: The results showed as for Table 3 that the daily intakes arsenic from aquatic products of various residents was different in Guangzhou. The daily intakes arsenic from aquatic products of high income groups was highest in all groups and the second group was standard man. The daily intakes arsenic from aquatic products of child was lowest in all groups. The daily intakes arsenic from aquatic products of various income groups was significant different which the higher income groups had the higher daily intakes arsenic. The main reason was the dietary intakes different of various types of aquatic products to various residents.

Food and Agriculture Organization of United Nations (FAO) /WHO combine with Codex Committee announce that the Tolerance Daily Maxium Intake (TDMI) of total arsenic is 50 μg/kg Body Weight (BW) and calculate the intake is 3000 μg/day of the standard body weight 60 kg. WHO also recommends Provisional Tolerable Weekly Intake (PTWI) of total arsenic is 15 g/kg BW, base on the standard man. The Arsenic Acceptable Daily Intake (ADI) of adult is about 0.128 mg/day (Tsuda *et al*., 1995a; Badal and Kazuo, 2002). The daily dietary total arsenic intake of America, Britain, Canada, Spain, Japan, France and Australia was 58.1, 120, 59.2, 286, 280, 36.9 and 62.3-80.4 g/day, respectively (Li *et al*., 2006; Egan *et al*., 2002; Ysart *et al*., 2000; Dabeka and McKenzie, 1995; Urieta *et al*., 1996; Tsuda *et al*., 1995b; Lee *et al*., 2006). The total arsenic intake from aquatic products of various residents was only a small proportion of the safety standard in this study.

The various types of aquatic products had different contribution to dietary intakes of arsenic. The freshwater fish had the largest contribution to the daily intakes arsenic from aquatic products in all type aquatic products, accounted for around 50%. Thus, freshwater fish was the main way of arsenic intake from aquatic products of residents in Guangzhou. Harry studied the daily dietary total arsenic intake in Belgium and found fish was the important source of arsenic (Robberecht *et al*., 2001). Recent studies have shown that even low concentrations of arsenic impair neurological function, particularly in children (Christina and Andrea, 2014). Although there was no human arsenic poisoning occurrence in recent years in Guangzhou, the high level of the daily arsenic intake suggested that the potential risk of arsenic poisoning for local residents, especially high income groups and the child might be a matter of concern.

CONCLUSION

- The total arsenic concentration was different in various marketed aquatic products. The range of concentration in aquatic products was 0.0075-1.2017 mg/kg and the mean arsenic concentration of aquatic products was 0.2022 mg/kg. The highest concentration aquatic product was crustacean in all types of aquatic products. The order of arsenic concentration of various aquatic production groups was crustacean> mollusk fish> saltwater fish>freshwater fish.
- The range of daily intakes arsenic from aquatic products was 5.96-11.85 μg/day. The daily dietary intakes arsenic from aquatic products of the child was lowest and the high income group was highest in all groups.

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