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Science of the Total Environment 361 (2006) 57–66

Science of the Total Environment
An International Journal for Scientific Research into the Environment and its Relationship with Humankind

www.elsevier.com/locate/scitotenv

Assessing the human health risks from exposure of inorganic arsenic through oyster (*Crassostrea gigas*) consumption in Taiwan

Chen-Wuing Liu [a,*], Ching-Ping Liang [b], Feng Mei Huang [a], Yu-Mei Hsueh [c]

[a] *Department of Bioenvironmental Systems Engineering, National Taiwan University, Taipei 106, Taiwan, ROC*
[b] *Department of Environmental Engineering and Sanitation, Foo-Yin University, Kaoshiung 831, Taiwan, ROC*
[c] *Department of Public Health, School of Medicine, Taipei Medical University, Taipei 110, Taiwan, ROC*

Received 30 September 2004
Available online 24 August 2005

Abstract

This study estimated the human health risk associated with ingesting inorganic arsenic through consumption of farmed oysters in Taiwan. Two hundred fifty-four samples of oyster (*Crassostrea gigas*) were collected from four townships in southwest coastal areas, where 90% of Taiwan's oysters are produced. The concentrations of total arsenic and arsenic species including As(V), As(III), monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA) were analyzed. The analytical results reveal that the ratio of mean concentration among the four townships of inorganic As to total concentration of As in oysters is approximately 1.64%. The mean concentrations of As(III) and As(V) in oysters from the four townships range from 0.071 to 0.145 μg/g, and 0.032 to 0.062 μg/g respectively. The estimated target cancer risks (*TR*), based on a 95% occurrence probability from ingesting inorganic As by consuming oysters at a rate of 18.6–56 g/day, range from 1.26×10^{-5} to 3.82×10^{-5}. The probabilities of TR fell within the range 10^{-6}–10^{-4}, suggesting that inorganic As uptake from farmed oysters is associated with a potential cancer risk. Moreover, a target hazard quotient (*THQ*) was used to evaluate the non-carcinogenic risk associated with ingesting inorganic As through oyster consumption at a rate of 18.6–56 g/day. The THQ values based on a 95% probability of exposure range from 0.071 to 0.214. All THQ values are below unity, indicating that farmed oyster consumption contributes only a little to the non-carcinogenic risk. Based on the estimation of the TR model, an ingestion rate of 1.6 g/day is recommended to meet the 95th percentile of carcinogenic risk, 10^{-6}, for exposure to inorganic As through the consumption of oysters in Taiwan.

Keywords: Oyster; Inorganic arsenic; Health risk assessment

* Corresponding author. Tel.: +886 2 2362 6480; fax: +886 2 2363 9557.
E-mail address: lcw@gwater.agec.ntu.edu.tw (C.-W. Liu).

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doi:10.1016/j.scitotenv.2005.06.005

1. Introduction

Arsenic is a ubiquitous element that is the 20th most abundant in the earth's crust, the 14th most abundant in seawater, and the 12th most abundant in the human body. Humans are exposed to arsenic from various sources, including food, water, air and soil, among others. Previous studies have found that exposure to arsenic may lead to cancers of the liver, kidney, bladder, prostate, lymphoid tissue, skin, colon, lung and nasal cavity (Chen et al., 1992); it may also cause blackfoot disease (BFD) (Ch'i and Blackwell, 1968), ischemic heart disease (Hsueh et al., 1998), hyperpigmentation, hyperkeratosis, diabetes (Tseng et al., 2000), meningioma and other detrimental effects on health (Chan and Huff, 1997). The International Agency for Research on Cancer (IARC) has classified arsenic as a group 1 carcinogen, meaning that sufficient evidence of human carcinogenity exists (IARC, 1980).

BFD is an endemic peripheral vascular disease that is frequently observed among inhabitants in a small coastal area in the southwest of Taiwan, where water in deep wells contains a high concentration of arsenic. Chen et al. (1994) reported total arsenic levels in well water from 470 to 897 μg/l in the hyperendemic BFD area in southwestern Taiwan. Ninety-five percent of all As is inorganic (As(III) and As(V)), and the predominant arsenic species is As(III). The inhabitants have not drunk well water since many epidemiological studies showed that exposure to arsenic was strongly associated with BFD (Chen et al., 1988, 2001). However, groundwater is still utilized in aquaculture (Huang et al., 2003).

Arsenic is present in various chemical forms, all of which can be easily absorbed by aquatic organisms. Arsenic can also accumulate in animal and human food chains (Cullen and Reimer, 1989; Edmonds et al., 1997). Humans are exposed to As from various sources, such as food, water, air and soil; food is the major source of As to which humans are exposed. The U.S. food and Drug Administration (U.S. FDA, 1993) indicated that fish and other seafood account for 90% of the total As exposure.

Suhendrayatna et al. (2002) also found that freshwater organisms accumulated and transformed arsenic in their bodies. Approximately 90% of accumulated arsenic was depurated to water. Several arsenic species are present in marine organisms; they include arsenobetaine, arsenocholine, monomethylarsonic acid (MMA), dimethylarsinic acid (DMA), As(V) and As(III) (Edmonds and Francesconi, 1987, 1993; Edmonds et al., 1997; Franceskoni and Kuehnelt, 2002). Arsenical toxicity, determined from the prevalence of carcinogenesis and vascular disorders in an earlier arsenic endemic of southwest coast of Taiwan, follows the order MMA(III)>As(III)>As(V)>MMA(V)=DMA(V) (Lin et al., 1998). Inorganic arsenic species are more toxic than methyl arsenic species. The U.S. EPA uses inorganic arsenic uptake by various seafood to determine the potential risk to human health (U.S. EPA, 1989, 2004).

Oysters are the most popular shellfish in Taiwan and the major farmed areas are off of the coast of southwest Taiwan, so the As content in oysters has received much attention. Han et al. (1998, 2000) used a deterministic risk analysis method to estimate the potential health risks from consuming metals in seafood in Taiwan. Ingesting inorganic arsenic through oyster consumption was associated with a high cancer risk of 5.14×10^{-4}. Guo (2002) critically examined the work of Han et al. (1998) and concluded that insufficient and improper data and assumptions were used had been responsible for unrealistic results in the latter work.

In this study, inorganic As contents in oysters were comprehensively surveyed to estimate the health risks associated with ingesting inorganic arsenic by consuming oysters. Monthly oyster samples were collected from four townships that supply 90% of the Taiwanese oyster market, from January to December 2002 (over 12 months). Inorganic As concentrations in oysters were directly measured. The carcinogenic and non-carcinogenic risks to health were evaluated probabilistically which accounted the uncertainty in the risk associated with the concentration of inorganic arsenic in oyster. The results were compared with those of Han et al. (1998) to ensure the validity of the assessed health risk obtained herein.

2. Materials and methods

2.1. Sample collection and analysis

Local fishermen harvest cultured oysters based on their shell size. When the oyster shell reaches 4 cm in

diameter and a wet weight of over 10 g, the oyster is ready to be harvested. Oyster (*Crassostrea gigas*) samples were collected from southwestern offshore aquaculture farms in Wangkung, Tungshih, Putai and Anpin — four townships that produce 90% of the oysters in Taiwan (Fig. 1). Oyster samples were purchased monthly at two locations in each township and three oyster samples from each location were analyzed to determine their total As and As species contents. However, purchases were not made in January, February and March in Wangkung; in February or March in Putai, or in January or February in Tungshih. The total number of oyster samples was 254. Oyster samples were frozen while they were transported from the field to the laboratory and stored at −20 °C until they were dissected.

A portion of the homogenized samples were freeze-dried for 36 h and prepared to analyze the total content of arsenic and arsenic species. About 0.5 g of homogenized freeze-dried samples and 25 ml of 65% nitric acid were added to a flask. They were boiled and decanted to allow gases to pass through a condenser and digested for 12 h until the solution was clear. Total arsenic was analyzed using an electro-thermal atomic absorption spectrometer (AAS) (AA100 Perkin-Elmer Shelton, USA), and a hydride generator (HG) (FIAS 400 Perkin-Elmer Shelton, USA). A 0.5% $NaBH_4$ in 0.25% NaOH

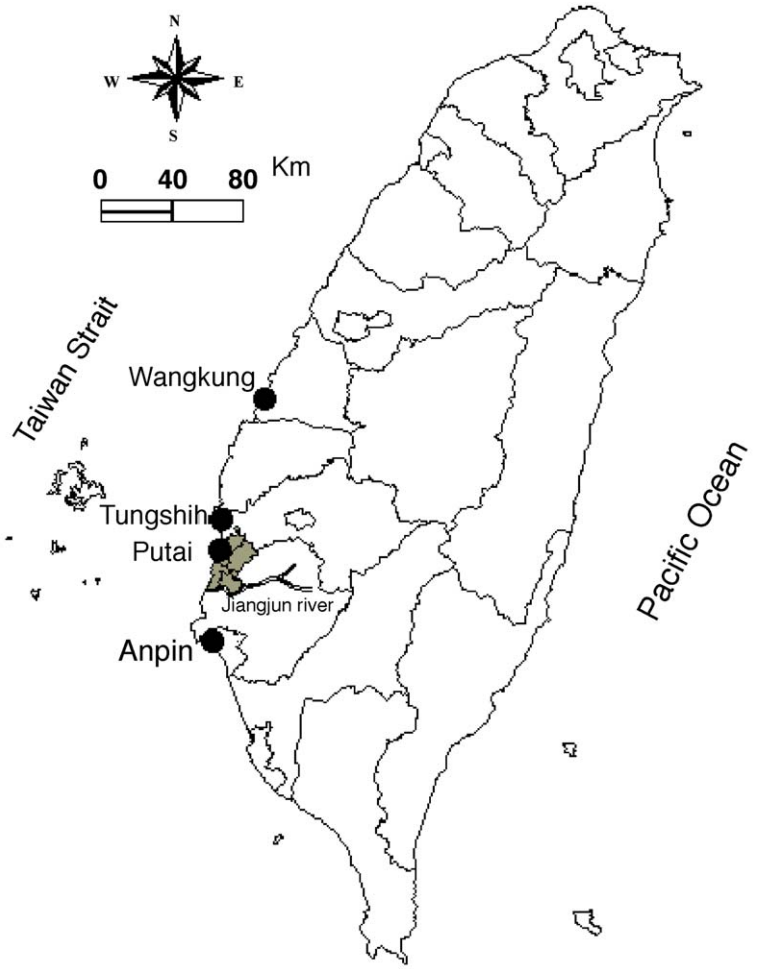

Fig. 1. Oyster sampling locations. The gray region represents the occurrence area of the blackfoot disease.

and 1N HCl were added to 200 μl of a digested sample to reduce the arsenic to arsine. Then, the total arsenic concentrations were determined by HG/AAS.

1.0~1.5 g of freeze-dried oyster and 150 ml of methanol/water solution (1/1, v/v) were placed into a Soxhlet extraction apparatus, modified from that described by Gomez-Ariza et al. (2000) and extracted for 16 h. A methanol extraction tube was designed to remove the methanol when the extraction was complete. After the methanol was removed, the extract was freeze-dried to a powder and re-dissolved in 10 ml of de-ionized water. The re-dissolved liquids were purified by filtering through C_{18} cartridges. A high-performance liquid chromatograph, HPLC (Hitachi 7110, Naka, Japan), equipped with an anion column (Machey-Nagel, Nucleosil, 10 μm, 250 × 4.6 mm) and connected to HG/AAS, was used to separate As(III), As(V), MMA and DMA. The procedures for analyzing As(III), As(V), MMA and DMA followed closely those in our previous study (Huang et al., 2003). Fig. 2 summarizes the method for analyzing total arsenic and arsenic species.

The accuracy of the procedure was validated by the analysis of the standard reference material (SRM) BCR 627 tuna tissue. Total arsenic and DMA concentrations of SRM were 4.9 ± 0.2 mg/kg and 1.8 ± 0.1 μmol/kg, respectively, which values were consistent with the certified values of 4.8 ± 0.3 mg/kg and 2.0 ± 0.3 μmole/kg, respectively. The detection limits of total arsenic, As(III), As(V), MMA and DMA were 0.2, 0.4, 0.2, 0.4 and 0.3 μg/1, respectively. Samples were spiked with arsenic species to calculate the recovery rate in every extraction step and laboratory procedure. The extraction recovery rates of As(III), As(V), MMA and DMA, were 102.7 ± 4.7%, 104.1 ± 6.8%, 104.7 ± 6.5% and 98.0 ± 7.1%, respectively. The laboratory procedure recovery rates of total As, As(III), As(V), MMA, and DMA were 103.2 ± 7.1%, 100.7 ± 3.8%, 97.2 ± 4.0%, 104.9 ± 4.6% and 97.2 ± 4.0%, respectively. The coefficient of variation was used to test the reliability and was less than 5% for all experiments.

2.2. Target cancer and non-cancer risk assessment

Risk assessments are based on assumptions. Assumptions should be clearly stated. They should be carefully validated. Real-life situations should be taken into account to prevent confusion and consequent adverse societal impact.

The Risk Assessment Forum (U.S. EPA, 1988) re-evaluated the carcinogenicity associated with ingesting inorganic arsenic. The U.S. EPA Region III Risk-Based Concentration Table (U.S. EPA, 2004) present methods for estimating the target cancer risk (TR) and the non-cancer risk (THQ). The risk associated with the carcinogenic effects of inorganic arsenic is expressed as the excess probability of contracting cancer over a lifetime of 70 years. The equation

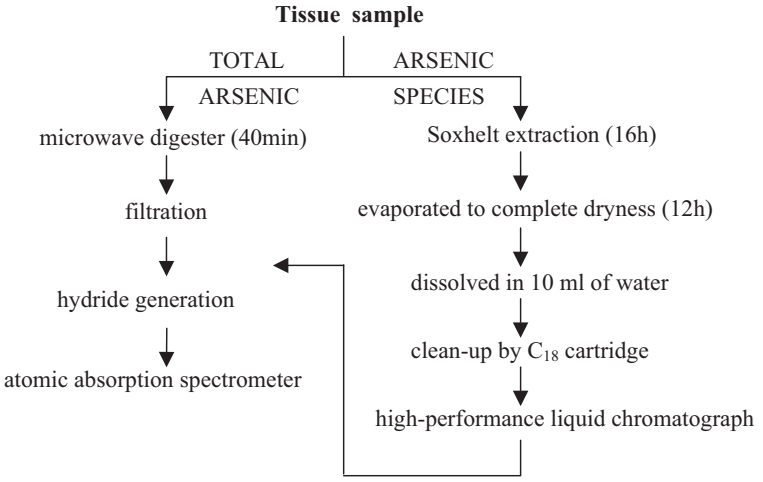

Fig. 2. Analytical method for total arsenic and arsenic species.

used for estimating the target cancer risk (lifetime cancer risk) is as follows (U.S. EPA, 1989),

$$TR = \frac{EFr \times ED_{tot} \times SFI \times MCS_{inorg} \times CPS_o}{BW_a \times AT_c} \times 10^{-3} \quad (1)$$

where TR represents the target cancer risk or the risk of cancer over a lifetime; EFr is the exposure frequency (days/year); ED_{tot} is the exposure duration (year); SFI is the mass of oyster ingested (g/day); MCS_{inorg} is the concentration of inorganic arsenic species in the edible portion of oyster (μg/g wet weight); CPS_o is the oral carcinogenic potency slope of inorganic arsenic (mg/kg/day); BW_a is the body weight of a Taiwanese adult; AT_c is the averaging time for carcinogens (days), and 10^{-3} is the unit conversion factor.

The non-cancer risk was estimated using the hazard quotient method, defined as follows (U.S. EPA, 1989).

$$THQ = \frac{EFr \times ED_{tot} \times SFI \times MCS_{inorg}}{R_fD \times BW_a \times AT_n} \times 10^{-3} \quad (2)$$

where THQ is the target hazard quotient (dimensionless); R_fD, is the oral reference dose (mg/kg/day); AT_n is the averaging time for non-carcinogens (day), and 10^{-3} is the unit conversion factor.

The exposure duration is 350 days/year for 30 years. An averaging time of 365 days/year for 70 years (AT_c=25,550 days) was used to characterize life time exposure for calculating cancer risk. An averaging time of 365 days/year for 30 years (AT_n=10,950 days) was used to characterize non-cancer risk. The cancer slope factor and the reference dose for ingested inorganic As are 1.50 $(mg/kg/day)^{-1}$ and 3×10^{-4} mg/kg/day, respectively, from the U.S. EPA IRIS data base (U.S. EPA, 2003). The average body weight of a Taiwanese adult is 60.5 kg, according to statistical data on 19–65 year old residents in Taiwan (http://www.doh.gov.tw/NewVersion/index.asp). Based on the U.S. EPA (1989) guideline, the ingested dose was assumed to equal the absorbed contaminated dose and that cooking does not affect the contaminants. Additionally, many factors modify the body burden of arsenic, after oysters have been consumed. Therefore, the measurement of arsenic in body fluid is critical to the determination of the health risk from such consumption. This study did not analyze body burden, and the effect of this omission on the estimated human health risk should be noted.

The input parameters in the health risk model have associated uncertainty (Lipton and Gillett, 1991). A range of oyster consumption rates, 18.6–56 g/day, was adopted to reduce the parametric uncertainty. Moreover, MCS_{inorg} in Eqs. (1) and (2) was treated probabilistically. @Risk (Version4.5, Professional Edition, Palisade Corp., USA) software was used to analyze measurements of the concentrations of inorganic arsenic species in oyster and estimated distribution parameters for inorganic arsenic species. A chi-square test was conducted to determine the probability distributions that best fit MCS_{inorg}. The Monte Carlo method was used to generate the distribution of inorganic arsenic in oysters from the determined best-fit distributions of concentrations of inorganic arsenic. The determined probability distribution was employed to describe the uncertainty of inorganic arsenic species in oyster. The estimated health risk associated with inorganic As consumed in oysters was expressed as a probability distribution, and the uncertainty in the risk associated with the concentrations of inorganic arsenic in the edible portion of oyster (MCS_{inorg}) was thus accounted for. Other parameters such as the body weight of Taiwanese adult were not treated probabilistically. The objective of this study is to evaluate the health risk from ingesting inorganic arsenic through oyster consumption. If all the parameters were inputted as probability distributions, then the results of the assessment would not allow the individual effects of each parameter to be determined easily and blue the focus of the study. An acceptable risk distribution was defined by constraints on percentiles. The lower end of the range of an acceptable distribution is defined by requiring that the 95th percentile of the risk distribution that must be equal to or lower than 10^{-6} for carcinogens and equal to or lower than unity for non-carcinogens.

3. Results and discussion

Table 1 presents the monthly measured total As concentration in oysters collected from the four townships. The highest and the lowest total As concentrations in four townships were 22.90 ± 7.14 and 4.22 ± 0.38 μg/g (mean ± SD). These were measured

Table 1
Monthly measured total and inorganic As concentrations (μg/g) in oysters from Anpin, Putai, Tungshih and Wangkung

Month	Total As concentration (mean ± SD)					Inorganic As concentration (mean)	
	Anpin	Putai	Tungshih	Wangkung	4 townships average (A)	4 townships average (B)	(B)/(A)%
1	12.39 ± 1.24	11.66 ± 0.60	–[a]	–	12.03 ± 0.52	0.293	2.433
2	4.22 ± 0.38	–	–	–	4.22 ± 0.38	0.111	2.639
3	7.60 ± 0.73	–	6.05 ± 0.44	7.55 ± 1.25	7.07 ± 0.88	0.086	1.219
4	8.31 ± 0.43	22.90 ± 7.41	9.95 ± 1.03	–	13.49 ± 8.24	0.289	2.14
5	7.15 ± 1.08	8.22 ± 0.44	8.95 ± 0.69	10.53 ± 0.42	9.01 ± 1.06	0.233	2.582
6	8.51 ± 2.90	11.72 ± 0.67	10.37 ± 0.96	10.28 ± 1.39	9.89 ± 1.93	0.138	1.4
7	7.50 ± 0.51	7.83 ± 1.30	8.55 ± 0.94	9.11 ± 1.30	8.51 ± 0.52	0.113	1.334
8	8.37 ± 0.33	10.12 ± 2.84	8.63 ± 0.79	11.39 ± 0.56	9.42 ± 1.70	0.085	0.902
9	8.12 ± 0.24	8.53 ± 1.29	9.44 ± 2.72	10.23 ± 1.85	9.15 ± 0.86	0.089	0.972
10	6.77 ± 1.28	8.14 ± 0.36	9.53 ± 2.25	13.24 ± 1.65	9.74 ± 2.41	0.095	0.971
11	7.62 ± 1.64	10.37 ± 1.06	7.07 ± 1.78	7.43 ± 0.95	8.13 ± 1.51	0.105	1.296
12	7.38 ± 0.49	7.32 ± 2.96	8.63 ± 0.65	8.19 ± 0.99	7.88 ± 0.63	0.138	1.75
12 month average	7.90 ± 1.80	10.68 ± 4.57	8.71 ± 1.30	9.78 ± 1.90	9.09 ± 3.08	0.148	1.636

–[a] Indicates no sample.

in April in Putai and in February in Anpin, respectively. The spatial concentration distributions (mean ± SD) of total As concentration in oysters were 7.90 ± 1.80, 10.68 ± 4.57, 8.71 ± 1.30 and 9.78 ± 1.90 μg/g in Anpin, Putai, Tungshih and Wangkung, respectively (Table 1). Putai had the highest mean total As concentration whereas Anpin had the lowest. The result of the Scheffe test (Brace et al., 2000) indicates that the concentration of total As of oyster in Putai and Wankung significantly exceeds that in Anpin.

The seasonal concentration distributions (mean ± SD) of total As in oyster are 9.32 ± 4.69, 9.58 ± 1.90, 8.85 ± 2.29 and 8.46 ± 2.61 in spring (March–May), summer (June–August), fall (September–November) and winter (December–February), respectively (Table 2). The results of the Scheffe test show the total As concentration in oysters does not vary significantly among the four seasons ($p > 0.005$).

Sanchez-Rodas et al. (2002) analyzed the same species of oyster (*C. gigas*) farmed off the Atlantic coast of Spain and showed that the total As concentration (mean ± SD) in oyster was 17.24 ± 0.25 μg/g. Vilano and Rubio (2001) also analyzed the same species of oyster farmed in northwest Spain. They found a total As concentration (mean ± SD) in oyster of 9.74 ± 0.37 μg/g. Kohlmeyer et al. (2002) examined the same species of oyster farmed in the Arcachon bay of France and found a total As concentration (mean ± SD) of 26.7 ± 0.5 μg/g.

The measured total As concentrations in oysters in the above studies all exceed the value herein of 9.09 ± 3.08 μg/g. In this work, the total As concentra-

Table 2
Seasonal concentration distribution of As species in oyster

As species	As species concentration (mean ± SD (μg/g))			
	Spring (60)[a]	Summer (72)	Fall (72)	Winter (42)
As(III)	0.125 ± 0.169	0.071 ± 0.048	0.062 ± 0.053	0.107 ± 0.077
As(V)	0.087 ± 0.135	0.046 ± 0.053	0.038 ± 0.040	0.055 ± 0.052
MMA	0.124 ± 0.163	0.065 ± 0.059	0.099 ± 0.132	0.052 ± 0.065
DMA	0.475 ± 0.411	0.385 ± 0.277	0.305 ± 0.261	0.444 ± 0.247
Total As	9.323 ± 4.687	9.576 ± 1.899	8.852 ± 2.290	8.456 ± 2.601
Inorganic As[b]/Total As (%)	2.27	1.22	1.13	1.92

[a] The number inside the parentheses denotes the sample number.
[b] Inorganic As is the summation of As(III) and As(V).

Table 3
Concentration distribution of As species in oyster of four townships

Arsenic species	As species concentration (mean ± SD (μg/g))				
	Anpin (72)[a]	Putai (60)	Tungshih (60)	Wangkung (54)	4 township average (246)
As(III)	0.073 ± 0.054	0.145 ± 0.151	0.084 ± 0.034	0.071 ± 0.038	0.091 ± 0.104
As(V)	0.057 ± 0.067	0.062 ± 0.032	0.044 ± 0.018	0.032 ± 0.016	0.057 ± 0.083
MMA	0.084 ± 0.095	0.106 ± 0.066	0.068 ± 0.032	0.050 ± 0.030	0.084 ± 0.112
DMA	0.272 ± 0.195	0.443 ± 0.371	0.446 ± 0.096	0.480 ± 0.075	0.480 ± 0.313
Total As	7.90 ± 1.80	10.68 ± 4.57	8.71 ± 1.30	9.78 ± 1.90	9.09 ± 3.08
Inorganic As[b]/ Total As (%)	1.65	1.94	1.47	1.05	1.64

[a] The number inside the parenthesis denotes the sample number.
[b] Inorganic As is the summation of As(III) and As(V).

tions in oysters from the four townships varied significantly. A spatial variation in total As concentration in oysters is clearly evident.

Han et al. (1997, 1998) measured the total As concentrations in oysters in Putai, finding values of 12.3–21.4 μg/g in 1997 with a geometric mean of 18.7 μg/g, and 3.15–7 μg/g in 1998 with a geometric mean of 4.86 μg/g. The total arsenic concentration of oyster in Putai in this study ranged from 3.11 to 33.37 μg/g with a geometric mean of 9.99 μg/g, suggesting that an annual temporal effect may apply. However, the seasonal variation of the total As concentration in oysters was statistically insignificant.

The annual average inorganic As was found to constitute 1.64% of the total As in oyster (Table 1). The highest and lowest percentages were 2.64% and 0.90%, in February and August, respectively. The spatial inorganic As fractions in Anpin, Putai, Tungshih and Wangkung were 1.65, 1.94, 1.47 and 1.05%, respectively (Table 3). The inorganic As fractions in spring, summer, fall and winter were 2.27, 1.22, 1.13 and 1.92%, respectively (Table 2). Edmonds and Francesconi (1993) reported that 1.4% of the arsenic in oysters (*C. gigas*) from Japan was inorganic As. Kohlmeyer et al. (2002) showed 3% of the arsenic in oysters (*C. gigas*) from the northwest of Spain was inorganic. Our result of 1.64% is between these values for Japan and Spain.

Fig. 3 plots the determined goodness-of-fit of the lognormal probability function LN (mean ± SD) of inorganic As concentrations (μg/g dry weight), based on the measured arsenic concentrations in oysters from the oyster farms of the four townships. The goodness-of-fit of the lognormal probability functions of the inorganic As concentration (μg/g dry weight) in oysters from the four townships in the four seasons is also determined. These determined lognormal probability functions were used to assess the risks to human health. The 18.6 and 56 g/day oyster ingestion rates of adults in Taiwan were used. Notably, 18.6 g is the mass of oyster in a typical fried oyster pancake, which is commonly served as a Taiwanese snack. Table 4 shows that the 95th percentile target cancer risk from ingesting inorganic As by eating 18.6–56 g/day of oyster in Taiwan is 1.26×10^{-5}–3.82×10^{-5}. The 95th percentile cancer risks were 1.30×10^{-5}–4.30×10^{-5},

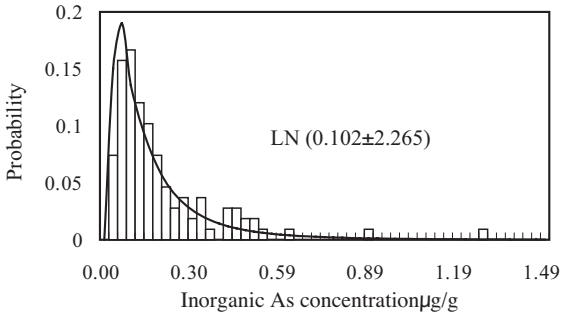

Fig. 3. Goodness-of-fit of lognormal distribution function LN (mean ± SD) of inorganic As concentration in oyster.

Table 4
Estimated target cancer risk (TR) and target hazard quotient of inorganic As due to consumption of oyster produced in the southwest Taiwan (THQ)

Cumulative probability percentile	Ingestion rate			
	18.6 g/d		56 g/d	
	TR	THQ	TR	THQ
5%	9.00E-07	0.005	2.80E-06	0.015
25%	2.18E-06	0.012	6.70E-06	0.037
50%	3.73E-06	0.021	1.11E-05	0.062
75%	6.21E-06	0.034	1.83E-05	0.102
95%	1.26E-05	0.071	3.82E-05	0.214

1.86×10^{-5}–5.6×10^{-5}, 7.92×10^{-6}–2.48×10^{-5} and 1.01×10^{-5}–3.03×10^{-5}, respectively, for Anpin, Putai, Tungshih and Wangkung. The estimated target cancer risks from ingesting inorganic As by consuming oysters all exceeded 10^{-6}, and the risk was highest in Putai. Notably, Putai is the area in the southwest of Taiwan that saw a hyperendemic blackfoot disease.

Han et al. (1998) assumed that inorganic As represented 10% of total As in oyster. The highest target cancer risk that they estimated was 5.10×10^{-4}, associated with a intake of a 19.3 μg/g dry weight As content oyster and consuming 139 g/day for 30 years. The mean cancer risk for consuming 18.6 g/day of oysters in Taiwan is 3.2×10^{-5} and the mean cancer risks in Anpin and Putai are 3.46×10^{-5} and 1.71×10^{-5}, respectively. Their results are higher than those in this study, because of the 10% inorganic As content, the high ingestion rate (139 g/day) and the failure to convert the dry weight As to wet weight As in oysters. The estimated cancer risk herein is obtained under conditions that more closely resemble reality. Additionally, Gagnon et al. (2004) estimated the target cancer risk of inorganic As consumed in shellfish harvested from the north shore of the St. Lawrence River's lower estuary. They used the same assessment method and estimated a TR of 3.75×10^{-5}, given a 17 g daily consumption rate of soft-shell clams (*Mya arenaria*). Their results are compatible with those in this study.

Table 4 presents the target hazard quotient of the 5, 25, 50, 75 and 95th percentiles of cumulative probability, from ingesting inorganic As by consuming 18.6 g/day of oysters. The 95th percentile target hazard quotients from ingesting inorganic As by consuming 18.6–56 g/day oyster in Taiwan were 0.071–0.214. The 95th percentile target hazard quotients were 0.075–0.026, 0.104–0.314, 0.047–0.139 and 0.056–0.169 in Anpin, Putai, Tungshih and Wangkung, respectively. The estimated target hazard quotients from ingesting inorganic As through oyster consumption were all below the acceptable safe value of unity. The highest target hazard quotient was found in Putai.

If a 95th percentile TR value of 10^{-6} is an acceptable risk, then the corresponding recommended daily ingestion rate of oyster can be calculated by Eq. (1). The inorganic As concentration (mean ± SD) 0.019 ± 0.017 μg/g (wet weight) in oyster is used, and the calculated daily ingestion rate of oyster is 1.6 g. This is about one tenth of the mass of oysters in a fried oyster pancake. Restated, people in Taiwan should eat no more than three fried oyster pancakes per month to have an acceptable health cancer risk (10^{-6}) from exposure to inorganic arsenic through oyster consumption.

4. Conclusions

This study directly measured the concentration of As species in oyster, to estimate the cancer and non-cancer health risks associated with exposure to As through oyster consumption in Taiwan. 254 oyster samples of oyster were collected monthly from four townships in the southwest coastal area in 2002. The work lasted for 12 months and the collected samples covered 90% of the oyster production area. Among the four townships the measured mean concentrations of total As, As(III) and As(V) were 9.09, 0.09 and 0.06 μg/g, respectively. The mean ratio of inorganic As to total As in oyster was 1.64%. The distribution of inorganic As in oyster was determined by the goodness-of-fit of a lognormal function. Using the best-fitted lognormal function of inorganic As concentration in oyster, the estimated 95th percentile cancer risks are 1.26×10^{-5} and 3.82×10^{-5} for ingestion of 18.6 g/day and 56 g/day oyster which all exceeds the acceptable cancer risk of 10^{-6}. Additionally, the estimated 95th percentile target hazard quotients were 0.071 and 0.214 for consumption of 18.6 and 56 g/day oyster. These values were both below unity and within the safety range for human health. Comprehensive data on inorganic As data in oyster were obtained herein and the estimated TR and THQ values adequately represent the health risk under realistic conditions. Based on the TR model estimation, an maximum ingestion rate of 1.6 g/day is recommended to ensure that people who ingest inorganic As by consuming oysters in Taiwan meet the 95th percentile carcinogenic risk, 10^{-6}.

Acknowledgements

We thank the National Science Council of the Republic of China for financially supporting this research under Contract No. NSC 91-2313-B-002-270.

We also thank Dr. Dipankar Chakraborti and Dr. Amitava Mukherjee for their constructive comments.

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