

# 行政院國家科學委員會專題研究計畫成果報告

## 無機砷於養殖魚類/螺貝類體內吸收,分布,排除及其風險評估之研究(3/3)

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### 摘要

第三年研究工作為運用機率風險分析方法，探討臺灣烏腳病地區養殖吳郭魚和豆仔魚之砷生物累積，並評估人類食入受污染魚體之暴露量範圍。此模式結合「生物累積機率模式」說明魚體之砷生物累積，及「人體健康暴露和風險模式」以說明人體攝入受污染魚體之危害商數(hazard quotient)和終生風險(lifetime risk)。結果顯示由無機砷估算第 95 個百分位(95<sup>th</sup> percentiles)之危害商數範圍：臺北市居民攝食率 10-70 g d<sup>-1</sup> 時，危害商數為 0.31-2.65；及烏腳病當地漁民攝食率 48-169 g d<sup>-1</sup> 時，危害商數範圍為 1.86-6.09。另外，由無機砷造成之第 95 個百分位(95<sup>th</sup> percentiles)人體潛在健康風險範圍：臺北市居民攝食率 10-70 g d<sup>-1</sup> 時，風險值為  $5.70 \times 10^{-5} - 5.25 \times 10^{-4}$ ；烏腳病當地漁民攝食率 48-169 g d<sup>-1</sup> 時，風險值為  $3.40 \times 10^{-4} - 1.12 \times 10^{-3}$ 。故由研究結果指出臺北市居民和當地漁民之砷暴露量所造成之風險有較保守的估計，並於研究中計算最大允許一標準單位之魚體內無機砷濃度，以第 95 個百分位數計算超過  $10^{-6}$  之終生風險值，可得當攝食率為 10-70 g d<sup>-1</sup> 時吳郭魚和豆仔魚最大允許無機砷濃度為 0.0019 - 0.0175 和 0.0023 - 0.0053  $\mu\text{g g}^{-1}$ ；攝食率為 48-169 g d<sup>-1</sup> 時，最大允許無機砷濃度為 0.0009 - 0.0029 和 0.0011 - 0.0013  $\mu\text{g g}^{-1}$ 。

關鍵詞：砷，吳郭魚，豆仔魚，風險評估，烏腳病。

### Abstract

This project carries out probabilistic risk analysis methods to quantify arsenic (As) bioaccumulation in cultured fish of tilapia (*Oreochromis mossambicus*) and large-scale mullet (*Liza macrolepis*) at blackfoot disease (BFD) area in Taiwan and to assess the range of exposures for the people whom eat the contaminated fish. The models implemented include a probabilistic bioaccumulation model to account for As accumulation in fish and a human health exposure and risk model that accounts for hazard quotient and lifetime risk for human consuming contaminated fish. Results demonstrate that the 95<sup>th</sup> percentiles of hazard quotient for inorganic As ranged from 0.31 - 2.65 for fish consumption rates of 10 - 70 g d<sup>-1</sup> for Taipei city residents, whereas 1.86 - 6.09 for subsistence fishers

in BFD area under 48 - 169 g d<sup>-1</sup> consumption rates. The highest 95<sup>th</sup> percentiles of potential health risk for inorganic As ranged from  $5.70 \times 10^{-5} - 5.25 \times 10^{-4}$  for eating fish harvested from the BFD area farms, under consumption rates of 10 - 70 g d<sup>-1</sup> for Taipei city residents, whereas  $3.40 \times 10^{-4} - 1.12 \times 10^{-3}$  for subsistence fishers under 48 - 169 g d<sup>-1</sup> consumption rates. These findings indicate that As exposure poses risks to residents and subsistence fishers, yet these results occur under highly conservative conditions. We calculate the maximum allowable inorganic As residues associated to a standard unit fish concentration based on the 95<sup>th</sup> percentile probability exceeding a  $10^{-6}$  lifetime risk, resulting the maximum target residues are 0.0019 - 0.0175 and 0.0023 - 0.0053  $\mu\text{g g}^{-1}$  for tilapia and large-scale mullet, respectively, under consumption rates of 10 - 70 g d<sup>-1</sup>, whereas 0.0009 - 0.0029 and 0.0011 - 0.0013  $\mu\text{g g}^{-1}$  for 48 - 169 g d<sup>-1</sup> consumption rates.

Keywords: Arsenic; Tilapia; Mullet; Risk assessment; Blackfoot disease.

### Introduction

Arsenic (As) is widespread in the environment as a consequent of both anthropogenic and natural processes. It is a ubiquitous but potentially toxic trace element. Inorganic as well as organic forms of As are present in the environment, and the former seems to be more toxic and slightly more accumulated in some freshwater aquatic species than the latter (Spehar *et al.* 1980). Trivalent As may show an adverse effect on aquatic biota and is considered more toxic than the inorganic pentavalent from (Hall and Burton 1982). Humans are exposed to arsenic (As) from many sources such as food, water, air and soil. US FDA (1993) in examining the food category indicated that fish and other seafood account for 90% of the total food As exposure with all other foods accounting for the remaining 10%. Donohue and Abernathy (1999) reported that the total As in marine fish, shellfish, and freshwater fish tissues are ranged from 0.19 - 65, 0.2 - 125.9, and 0.007 - 1.46  $\mu\text{g g}^{-1}$ , respectively.

Chen *et al.* (1985, 1986, 1988, 1992), in a series studies in Taiwan, indicated that an association between inorganic As exposure and cancer mortality from cancers for the lung, liver, and bladder has been documented among residents in the blackfoot disease

(BFD) area in that inorganic As levels in drinking water ranged from 0.01 – 1.752 mg L<sup>-1</sup>. BFD is a peripheral vascular disorder found in a limited area on the southwestern coastal area of Taiwan. The BFD area consists mainly of four towns, Putai, Yichu, Peimen and Hsuehchia, located at Chiayi and Tainan counties (Chen *et al.* 1980).

The cause of BFD is still unknown, but it generally attributed to the high concentrations of As found in the groundwater. Guo *et al.* (1994) reported that an association between inorganic As well-water concentrations and the incidence of urinary bladder and kidney cancer was demonstrated using data on 243 townships in Taiwan. Hsueh *et al.* (1995) indicated that multiple risk factors associated with arsenic-induced skin cancer based on epidemiological studies of residents from fishing communities of the BFD area in Taiwan. Chiou *et al.* (1995) reported that the internal cancer incidence for individuals with BFD in southwestern Taiwan was found to be increased compared to healthy residences of the same area in a 7-yr prospective study. Nowadays, most of the people living in these areas do not drink water from wells because tap water has been made available in this area; however, the groundwater is still used for aquaculture.

Lin *et al.* (2001), Singh (2001), and Liao *et al.* (2002) conducted a long-term investigation during 1998 – 2001 in BFD area indicated that As have been detected in many aquacultural ponds in that As concentrations in aquacultural waters are reported to range from 26.3±16 to 251.7 ±12.2 µg L<sup>-1</sup>, whereas whole-body burdens of As in cultured fish are ranged from 0.94±0.3 to 15.1±8.2 µg g<sup>-1</sup>. The results are much greater than the maximum contaminant level (MCL) for As in drinking water of 50 µg L<sup>-1</sup>.

Han *et al.* (1994, 1996, 1998) reported that the consumption of contaminated seafood has been as an important route of human exposure to heavy metals (As, Cu, Zn, Pb, Cd, Hg) in Taiwan in that oyster (*Crassostrea gigas*) and other seafood (e.g., tilapia, tuna, and shrimp) are the most popular seafood. Farming of tilapia (*Oreochromis mossambicus*) and large-scale mullet (*Liza macrolepis*) is a promising aquaculture in the BFD area because of high market value. The fish are fed with artificial bait, which does not contain As. These fish are maintained in the ponds for at least 8 months (from March to October) before they go to the marketplace. If waterborne As levels are elevated, toxicity can occur and have severe effects on the health of cultured fish, which will reduce market prices and cause closure of fish farms. Tilapia can also be used as a bioindicator for As bioaccumulation to study the accumulation and transformation of As in freshwater organisms (Suhendrayatna *et al.* 2002).

Currently, exposure estimates and subsequent human health and ecological risk projections usually assume a static and continuous exposure of an

represented by some descriptive statistics, such as the mean or maximum. Han *et al.* (1998) used a deterministic risk analysis method to estimate target hazard quotients and potential health risks for metals by consumption of seafood in Taiwan. Deterministic results, however, may hide significantly different levels of conservatism in relation to the uncertainty and variability present in each exposure parameter. Vermeire *et al.* (2001) pointed out that probabilistic modeling has received increasing support as a promising technique for characterizing uncertainty and variation in exposure estimates to environmental contaminants. To date, however, only a limited number of risk assessments regarding aquacultural management have incorporated probabilistic analyses. A predictive assessment is needed to evaluate the potential for As bioaccumulation, toxic effects to fish, and risks to human health (Reinert *et al.* 1991).

The purpose of this project is to propose a framework for risk assessment associated with As-contaminated aquacultural fish farms in developing As exposure estimates for tilapia and large-scale mullet in BFD area. The implications for human health risk estimates for people including city residents and subsistence fishers whom eat tilapia and large-scale mullet harvested from BFD area are also described.

## Materials and Methods

### Bioaccumulation Model

We used a first-order one-compartment model to describe uptake and elimination processes of fish exposed to As in an aquacultural pond and to calculate As body burden in fish over time. The first-order one-compartment model for the gain and loss of As accumulation in fish features constant biokinetic rates and constant water concentration. Accordingly, the dynamic behavior would be represented as

$$\frac{dC_f(t)}{dt} = k_1 C_w - k_2 C_f(t) \quad (1)$$

where  $C_f(t)$  is the time-dependent As concentration in fish (µg g<sup>-1</sup>),  $t$  is the time of exposure (d),  $C_w$  is the dissolved As concentration in water (µg g<sup>-1</sup>),  $k_1$  is the uptake rate constant from dissolved phase by fish (mL g<sup>-1</sup> d<sup>-1</sup>), and  $k_2$  is the depuration rate constant for As in fish (d<sup>-1</sup>).

We consider the steady-state condition in Eq. (1) and solve for  $C_f$  gives,

$$C_f = \frac{k_1}{k_2} C_w = \text{BCF} C_w \quad (2)$$

where  $\text{BCF} \equiv k_1/k_2 = C_f/C_w$  is the equilibrium bioconcentration factor (BCF) for fish (mL g<sup>-1</sup>). By incorporating distributions for input parameters, Eq. (2) can be run probabilistically.

**BCFs and Water Concentrations.** Of the variables used to estimate the distributions of As concentration in fish. BCF and  $C_w$  in Eq. (2) are

to develop probability distributions for BCFs and As concentrations in water. Data on As concentrations in pond water and fish tissue including gill, liver, muscle, intestine, and stomach were derived from the

$$\text{TR} = \frac{C_f \times \left( \text{CSF}_{\text{IRIS}} \left( \frac{\text{BW}}{70\text{kg}} \right)^{1/3} \right) \times \text{IR}_f \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}_c \times 10^3} \quad (3)$$

to develop probability distributions for BCFs and As concentrations in water. Data on As concentrations in pond water and fish tissue including gill, liver, muscle, intestine, and stomach were derived from the 1998 – 2001 field survey in BFD area by Singh (2001), Lin *et al.* (2001), and Liao *et al.* (2002). They chose three appropriate management practices fish farms for each sampling location. All cultured farms had similar feeding strategies. In this study, we choice Yichu, Hsuehchia, Peikangtzu, and Putai located at BFD area in southwestern costal area of Taiwan as our study sites in that fish farms in Yichu, Hsuehchia, Peikangtzu were cultured tilapia (*Oreochromis mossambicus*), whereas Putai was cultured large-scale mullet (*Liza macrolepis*). Minimum, mean, standard error, or maximum values of BCFs and water As concentrations were sorted to produce frequency distributions corresponding to each sampling site. A mathematical distribution was selected to express the range of variation and likelihood of BCF and  $C_w$  values within the range.

**Statistical Analysis.** The data were dividing into a minimum of ten bins as equally as possible. Absolute and relative frequencies were calculated and distributions were plotted using bin midpoints. We used the chi-square ( $\chi^2$ ) and the Kolmogorov-Smirnov (K-S) statistics (Zar 1999) to optimize the goodness-of-fit of distributions. We employed @RISK (Version 4.5, Professional Edition, Palisade Corp., USA) to analyze data and to estimate distribution parameters. The @RISK generated  $p$  values for the  $\chi^2$  statistics and provided critical values of  $D_{max}$  for the K-S statistics to estimate  $\alpha$  levels from 0.01 to 0.50. The selected distribution type and parameters were based on statistical criteria, comparisons of distribution parameters, and visual interpretation of histograms. US EPA (1997) in guiding principles for Monte Carlo analysis indicated that fit in the vicinity of expected values and in the tails were important criteria.

Finely *et al.* (1994) and Thompson *et al.* (2000) indicated that the lognormal distribution is often considered the default in environmental analysis. Distributions were fit to polled BCF data and the selected lognormal distributions had the acceptable  $\chi^2$  fit and K-S fit in that optimizations using either statistics yielded geometric mean (gm) and geometric standard deviation (gsd) expressing as LN(gm, gsd) (Figure 1). Water concentrations were also characterized by lognormal distributions by appropriately transforming from normal distributions for the mean with uncertainties characterized by standard error of the mean expressing as L(mean, sd) (Figure 1).

#### Human Health Exposure and Risk Model

The target cancer risk to adults is defined as

$$TR = \frac{C_f \times \left( CSF_{IRIS} \left( \frac{BW}{70kg} \right)^{1/3} \right) \times IR_f \times EF \times ED}{BW \times AT_c \times 10^3}, \quad (3)$$

where TR is the incremental individual lifetime cancer risk,  $CSF_{IRIS}$  is the oral carcinogenic slope factor from IRIS (Integrated Risk Information System, provided by US EPA) database ( $mg\ kg^{-1}\ d^{-1}$ ),  $IR_f$  is the annualized fish ingestion rate ( $g\ d^{-1}$ ),  $C_f$  is the As concentration in fish ( $\mu g\ g^{-1}$ ), EF is the exposure frequency (d/yr), ED is the exposure duration (yr),  $AT_c$  is the averaging time for carcinogens (d), BW is the body weight (kg), and  $10^3$  is the unit conversion factor.

The noncancer risk was estimated using the hazard quotient approach, defined as

$$HQ = \frac{C_f \times IR_f \times EF \times ED}{\left( RfD_{IRIS} \left( \frac{BW}{70kg} \right)^{1/3} \right) \times BW \times AT_{nc} \times 10^3}, \quad (4)$$

where HQ is the toxicity hazard quotient,  $RfD_{IRIS}$  is the oral reference dose from IRIS database ( $mg\ kg^{-1}\ d^{-1}$ ),  $AT_{nc}$  is the averaging time for noncarcinogens (d), and  $10^3$  is the unit conversion factor. We treated  $C_f$  and  $IR_f$  in Eqs. (3) and (4) probabilistically.

**Exposure Duration.** The outputs of the bioaccumulation model are predictions of As concentrations in tissue of an individual fish over time. The exposure duration is defined as the exposure frequency of 360 d/yr for 30 yr (i.e., 10,950 d). The averaging time and number of fish consumed are required to provide input for an estimate of human health risk from exposure through fish ingestion. An averaging time of 365 d/yr for 70 yr (i.e.,  $AT_c = 25,550d$ ) was used to characterize lifetime exposure for cancer risk calculation. An averaging time of 365 d/yr for 30 yr (i.e.,  $AT_{nc} = 10,950\ d$ ) was used in characterizing noncancer risk.

**Fish Ingestion.** Data on fish consumption patterns were adapted from two sources: (a) Han *et al.* (1998), which was based on a brief questionnaire about seafood consumption frequency and weeks of consumption for 850 residents in Taipei city and (b) Lin (unpublished work, 2002), which was based on a questionnaire on tilapia and large-scale mullet daily consumption rate for 57 subsistence fishers in BFD area. Han *et al.* (1998) provided data for fish ingestion rates for adult consumption of cultured fish in Taipei city of Taiwan. The fish ingestion rates ranged from 10 – 30 and 35 – 70  $g\ d^{-1}$  for 2 – 6 and 7 – 14 meals per week, respectively (Han *et al.* 1998). Lin (unpublished work, 2002) provided data on tilapia daily consumption rates for subsistence fishers in BFD area: 48 – 143 and 84 – 169  $g\ d^{-1}$  for 2 – 6 and 7 – 14 meals per week, respectively. We approximated these data using a lognormal distribution and were transformed appropriately to ensure the data did not differ from a normal distribution before parametric analysis. Results give fish ingestion rate distributions of LN(14.56, 2.05)

and LN(43.52, 1.87) for 2 – 6 and 7 – 14 meals per week, respectively, for Taipei city residents, whereas LN(163.07, 2.61) and LN(104.79, 1.75) for 2 – 6 and 7 – 14 meals per week, respectively, for subsistence fishers in BFD area. It was assumed in accordance with the US EPA (1989a) guideline that the ingested dose is equal to the absorbed contaminated dose and that cooking has no effect on the contaminants.

Schoof *et al.* (1999) and Donohue and Abernathy (1999) reported that the amount of inorganic As in seafood is ranged from < 3 – 7% of the total As. In this work, we assume inorganic As accounts for 5% of the total As in seafood.

**Body weight.** We used a 65 kg body weight for an average Taiwanese adult, as suggested by Han *et al.* (1998).

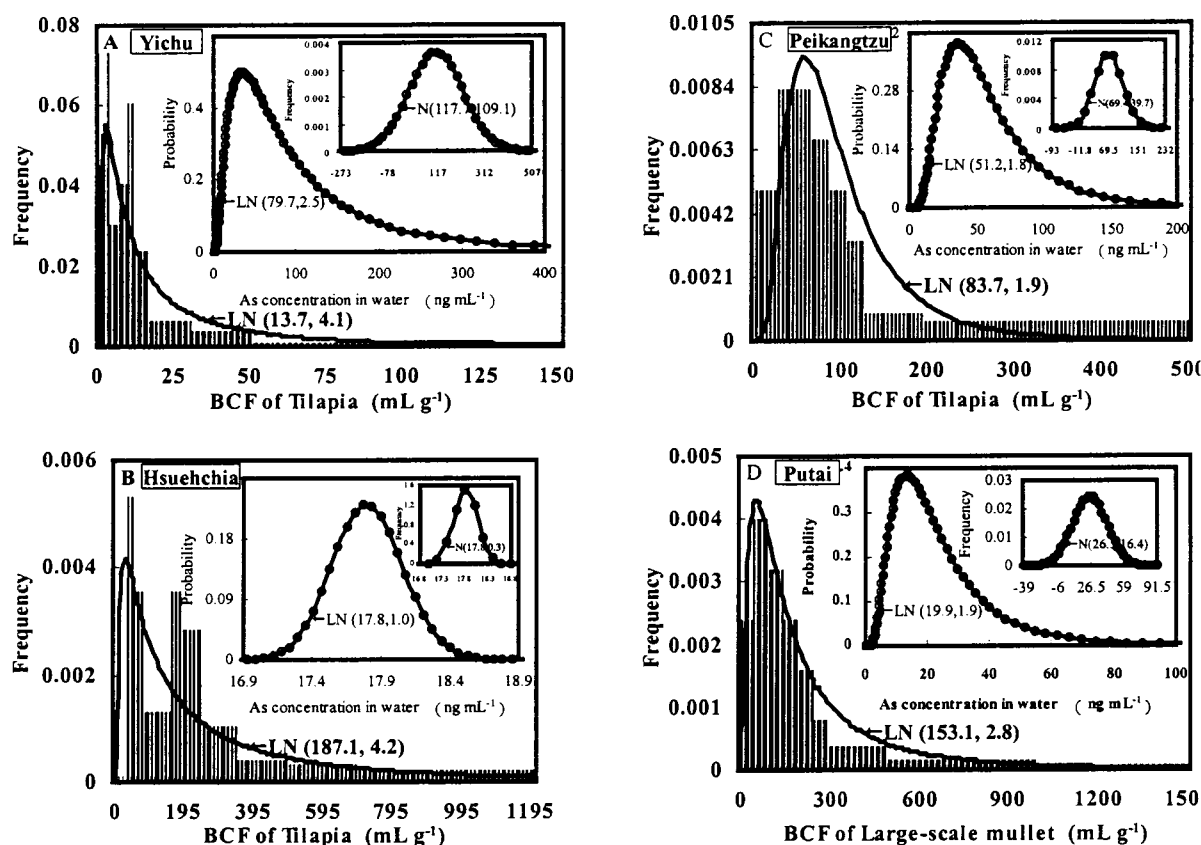


Fig. 1. Probability density functions of optimized lognormal distribution with geometric mean and geometric standard deviation as LN(gm, gsd) of BCFs and arsenic concentration in pond water for fish farms in (A) Yichu, (B) Hsuehchia, (C) Peikangtzu, and (D) Putai. The histograms of source data represented by frequency functions are also shown

**Toxicity Factors.** The cancer slope factor and reference dose for ingested inorganic arsenic are  $1.50 \text{ (mg kg}^{-1} \text{ d}^{-1})^{-1}$  and  $3 \times 10^{-4} \text{ mg kg}^{-1} \text{ d}^{-1}$ , respectively, provided by US EPA IRIS database (<http://www.epa.gov/iris>, 2001) and normalized to account for extrapolation to a different body weight from the standard of 70 kg (Eqs. (3) and (4)), as suggested in the *Exposure Factors Handbook* (USEPA 1997). These values are specified as point estimates following US EPA guidance (1989b).

**Acceptable risk distribution.** The acceptable risk distribution was assigned by constraints on percentiles. The lower end of the range of acceptable risk distribution is defined by a single constraint on the 95<sup>th</sup> percentile of risk distribution that must be equal or lower than  $10^{-6}$  for carcinogens and equal or lower than 1 for noncarcinogens.

### Simulation Scheme

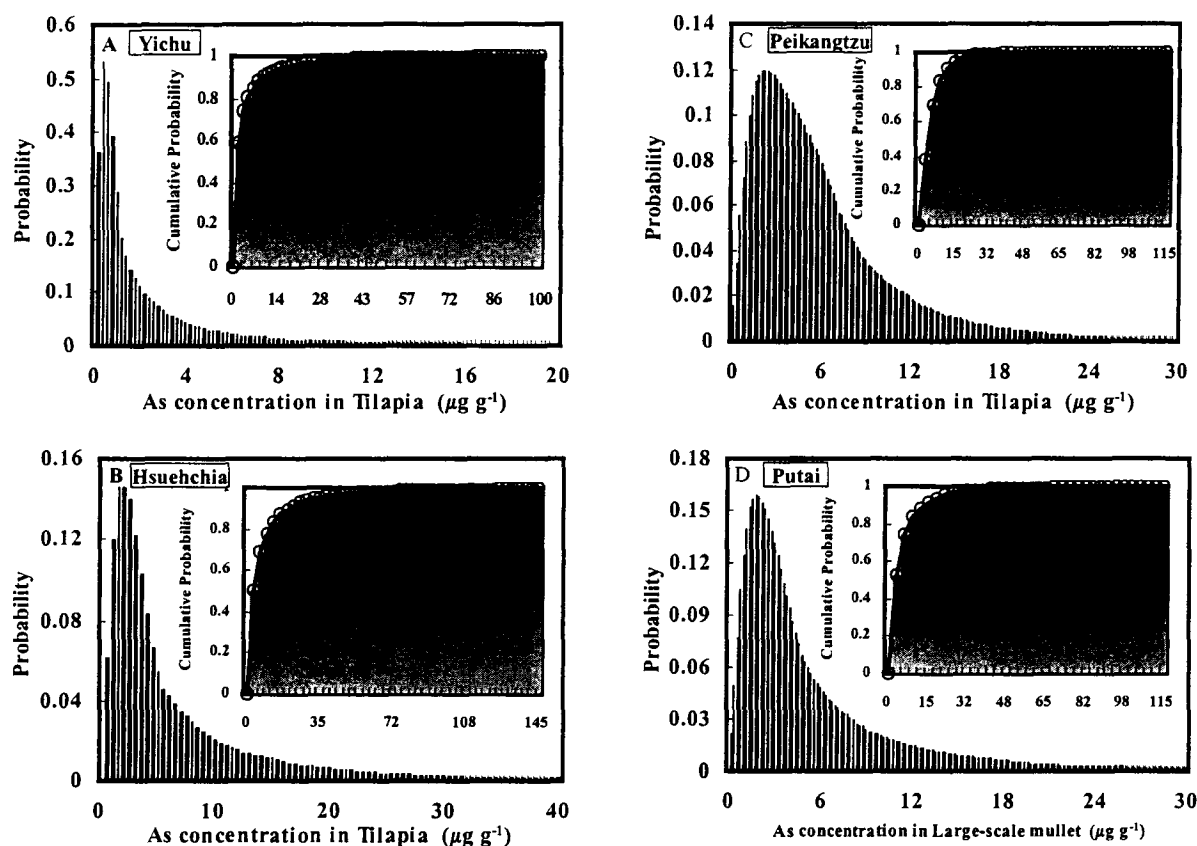
We are interested in the long-term equilibrium rather than the dynamics over a single growing season. We used Eq. (2) to predict As concentrations in cultured fish. Because the idea of the present model was to incorporate uncertainty into the model by selecting model parameters from lognormal probability distributions rather than experimentally derived values or field observations, we used a Monte Carlo technique to deal with the uncertainty (Vose 2000). Largely because of limitations in the data used to derive model parameters, inputs were assumed to be independently.

### Results and Discussion

Figure 2 illustrates the probability density functions (pdfs) and cumulative distribution

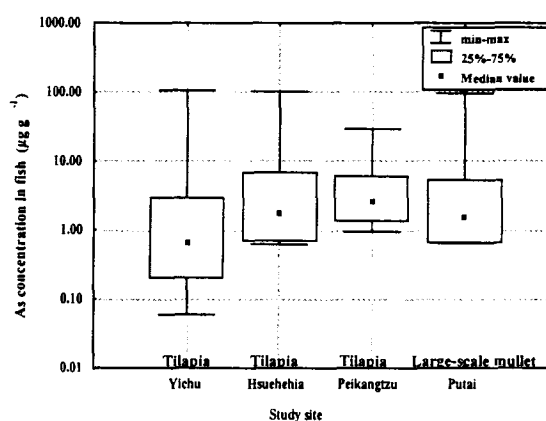
functions (cdfs) of As contents in tilapia and large-scale mullet from the fish farms in the BFD area. Probabilistic simulations of the bioaccumulation models produced skewed distributions of predicted As concentrations in fish. Percentile predictions of As contents in fish could be

determined from cdfs illustrated in Figure 2. Figure 3 shows box plots of interquartile and 50<sup>th</sup>-percentile predictions associated with whisker plots indicating measured minimum and maximum values of As concentrations in tilapia and large-scale mullet in the BFD area.



**Fig. 2.** Simulation results showing probability density functions and cumulative distribution functions of arsenic concentration in tilapia for fish farms in (A) Yichu, (B) Hsuehchia, (C) Peikangtzu, and large-scale mullet in (D) Putai.

Compared with the field observations, measured mean As concentrations in tilapia and large-scale mullet were larger than median estimates and all fall outside the interquartile range except in Yichu, yet all fall within the 5<sup>th</sup> and 95<sup>th</sup> percentile range (Figures 2 and 3). Relative to minimum and maximum field data, however, lower and upper probabilistic percentile predictions were more conservative. This is evidence that the modeling framework and the distributional parameters and assumptions in the model are appropriate for estimating bioaccumulation of As in cultured fish. The relative skewness and spread in modeled output varied between water and fish, distributions of As concentrations in fish were more skewed, with a long tail at higher concentrations (Figure 2). Measurements with minimum and maximum were less widely spaced, or less conservative, than the 5<sup>th</sup> and 95<sup>th</sup> percentile values of probabilistic output.



**Fig. 3.** Box and whisker plot representations of arsenic concentration in tilapia collected from fish farms in Yichu, Hsuehchia, and Peikangtzu, and in large-scale mullet collected from fish farms in Putai

Therefore, for As accumulation in fish, the BCFs or uptake/depuration rate constants of fish are the most influential variables.

Figures 4 and 5 compares hazard quotient (HQ) and target cancer risk (TR), respectively, for human consuming tilapia and large-scale mullet by Taipei city residents and subsistence fishers in the BFD area, respectively. The x-axis represents fish consumption rates along with fish farms in the BFD area in which the cultured fish goes to marketplace, whereas y-axis shows HQ and TR resulting from fish consumption by human under various meals per week. Under most regulatory programs, a HQ exceeding 1 and a TR between  $10^{-4}$  and  $10^{-6}$  indicate potential risk. Box and whisker plots represent the distribution of risks corresponding to the people lived in Taipei city and subsistence fishers whom eat the cultured fish harvested from fish farms in BFD area.

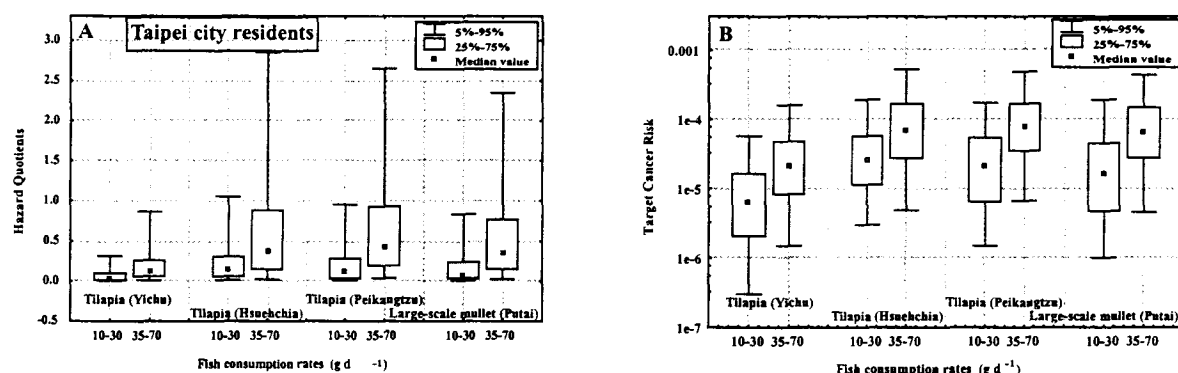


Fig. 4. Hazard quotients and target cancer risks for human consuming tilapia and large-scale mullet harvested from fish farms in the BFD area under different fish consumption rates for Taipei city residents. Box and whisker plots are used to represent the uncertainty in risk estimates for each ranged fish consumption rate

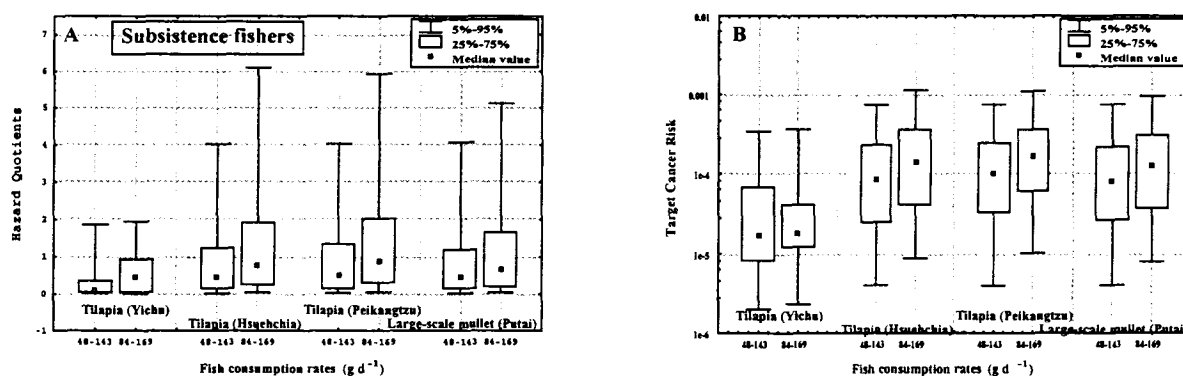


Fig. 5. Box and whisker plots representation of hazard quotients target cancer risks for human consuming tilapia and large-scale mullet for subsistence fishers in the BFD area for each ranged fish consumption rate

that HQ does not define a dose-response relationship, and hence its numerical value should not be regarded as a direct estimate of risk. Han *et al.* (1998) further indicated that cancer risk estimates for consumption of inorganic As in fish from the BFD area ranged between  $10^{-5}$  and  $10^{-4}$  for fish consumption rates of 10 – 70 g d<sup>-1</sup>, indicating high potential human health

risks. Figure 4 shows that for Taipei city residents, a 95% probability or less of experiencing a HQ less than 1 for daily consumption rate of 10 – 30 g d<sup>-1</sup>, indicating that these probability distributions are acceptable; whereas most of the HQs are larger than 1 for 35 – 70 g d<sup>-1</sup> fish consumption rate. All 95% probabilities of TR are larger than  $10^{-6}$ , indicating unacceptable probability distributions for Taipei city residents (Figure 4). For subsistence fishers in the BFD area, 95% probability HQs or TRs are larger than 1 or much fall outside the range of  $10^{-6}$  –  $10^{-4}$ , indicating high potential health risks (Figure 5). Han *et al.* (1998) reported that HQs caused by consuming fish containing As ranged from 0.136 – 0.340 for fish consumption rates of 10 – 70 g d<sup>-1</sup> in that they assumed inorganic As constitutes 10% of total arsenic in seafood. Han *et al.* (1998) also indicated

risks.

If compared with the acceptable 95<sup>th</sup> percentile probability of exceeding a  $10^{-6}$  TR and 1 HQ, we can calculate the maximum allowable fish residual level associated to a standard unit fish concentration. In doing so, the TR and HQ distributions associated to a unit fish concentration of inorganic As were rescaled

so that the 95<sup>th</sup> percentile is  $10^{-6}$  for carcinogens and 1 for noncarcinogens.

The calculated allowable fish residue is equal to  $f$  times the unit fish concentration where  $f_i = 10^{-6} / R_i^{95}$  for the  $i$ th carcinogen and  $f_j = 1 / HQ_j^{95}$  for  $j$ th noncarcinogen; where  $R^{95}$  being the 95<sup>th</sup> percentile of the TR distribution and  $HQ^{95}$  being the 95<sup>th</sup> percentile of the HQ distribution associated to a unit fish residue level. The maximum allowable residual concentrations in tilapia and large-scale mullet are 0.0019 – 0.0175 and 0.0023 – 0.0053  $\mu\text{g g}^{-1}$ , respectively, for Taipei city residents under consumption rates of 10 – 70  $\text{g d}^{-1}$ ; whereas 0.0009 – 0.0029  $\mu\text{g g}^{-1}$ , respectively, for subsistence fishers in the BFD area under consumption rates of 48 – 169  $\text{g d}^{-1}$ ; based on the 95<sup>th</sup> percentile probability exceeding a  $10^{-6}$  TR or 1 HQ (Table 1). Table 1 also indicates that the risks associated with exposure by consuming tilapia harvested from Hsuehchia fish farms in allowable residual concentrations have a greater likelihood of occurrence than the same risks associated with exposure to the other study sites.

**Table 1.** Calculated probabilistic maximum allowable fish residual levels for inorganic arsenic in BFD area in Taiwan

Study site	Fish	Fish consumption rate ( $\text{g d}^{-1}$ )	Probabilistic allowable fish residue ( $\mu\text{g g}^{-1}$ ) <sup>a</sup>	
			carcinogenic	noncarcinogenic
Yichu	Tilapia	10-30 <sup>b</sup>	0.0175	3.23
		35-70 <sup>b</sup>	0.0063	1.16
		48-143 <sup>c</sup>	0.0029	0.54
		84-169 <sup>c</sup>	0.0028	0.51
Hsuehchia	Tilapia	10-30	0.0052	0.95
		35-70	0.0019	0.35
		48-143	0.0014	0.25
		84-169	0.0009	0.16
Peikangtzu	Tilapia	10-30	0.0058	1.05
		35-70	0.0021	0.38
		48-143	0.0014	0.25
		84-169	0.0009	0.17
Putai	Large-scale mullet	10-30	0.0053	1.20
		35-70	0.0023	0.43
		48-143	0.0013	0.25
		84-169	0.0011	0.19

<sup>a</sup> A standard unit fish concentration ( $1 \mu\text{g g}^{-1}$ ) is considered.

<sup>b</sup> Ranged fish consumption rates for Taipei city residents (Han *et al.* 1998).

<sup>c</sup> Ranged fish consumption rates for subsistence fishers in BFD area (Lin, unpublished work, 2002).

This information implies that the mean value chosen in the deterministic bioaccumulation model for BCFs contribution to As accumulation in fish may not be sufficiently conservative: they will lead to target residual levels (see Table 1) associated with a probability of exceeding a  $10^{-6}$  TR or 1 HQ, higher than the threshold considered acceptable in the probabilistic context. For example, if the mean BCF value corresponding to the 75% percentile of this parameter distribution; that being so, the allowable concentrations for As in fish are strongly influenced by BCF, and the resulting 75% level of conservatism implied in the mean value is insufficient to ensure a 95% level of conservatism in the target risk value of  $10^{-6}$  calculated in the deterministic context.

In conclusion, this paper illustrates the use of a

simple bioaccumulation modeling in risk analysis. If used in a realistic fashion, it can more fully inform the decision-making process for the management of contaminated fish and can help support aquacultural water management decision by providing a quantitative expression of the confidence in risk estimates. The model could be also modified to incorporate additional complexities and numbers of sites and contamination profiles. The ability to use and interpret such models, however, is often limited by the state of knowledge concerning the spatial/temporal behavior of aquacultural ecosystems. Nevertheless, probabilistic treatment of the model parameters, coupled with sensitivity analyses, should provide a rigorous basis for making sound environmental decisions. With proper application of risk communication, we can increase human understanding of fish consumption strategies, and we can channel this legitimate concern into actions that will result in stricter water quality regulations. The end result of such action will be improved water quality, which will benefit the health of the fish and the health of the people whom eat them.

## References

- Balthis WL, Voit EO, Meaburn GM (1996) Setting prediction limits for mercury concentrations in fish having high bioaccumulation potential. *Environmetrics* 7:429-439
- Chen CJ, Chen CW, Wu MM, Kuo TT (1992) Cancer potential in liver, lung, bladder, and kidney due to ingested inorganic arsenic in drinking water. *Br J Cancer* 66:888-892
- Chen CJ, Chuang YC, Lin TM, Wu HY (1985) Malignant neoplasms among residents of a blackfood disease endemic area in Taiwan: high arsenic artesian well water and cancers. *Cancer Res* 45:5895-5899
- Chen CJ, Chuang YC, You SL, Wu HY (1986) A retrospective study on malignant neoplasms of bladder, lung, and liver in blackfood disease endemic area in Taiwan. *Cancer Res* 53:399-405
- Chen CJ, Wu MM, Kuo TL (1988) Arsenic and cancers. *Lancet* 1:414-415
- Chen CJ, Wu MM, Lee SS, Wang JD, Cheng SH, Wu HY (1980) Atherogenicity and carcinogenicity of high-arsenic well water: multiple risk factors and related malignant neoplasms of blackfood disease. *Arteriosclerosis* 8:452-460
- Chiou HY, Hsueh YM, Liaw KF, Horng MH, Pu YS, Lin JSN, Huang CH, Chen CJ (1995) Incidence of internal cancers and ingests inorganic arsenic: a seven-year follow-up study in Taiwan. *Cancer Res* 55:1296-1300
- Donohue JM, Abernathy CO (1999) Exposure to inorganic arsenic from fish and shellfish. In: Chappell WR, Abernathy CO, Calderon RL (eds) *Arsenic exposure and health effect*. Elsevier, Oxford, UK, pp89-98

- El-Shaarawi AH, Viverros R (1997) Inference about the mean in log-regression with environmental applications. *Environmetrics* 8:569-582
- Finley B, Proctor D, Scott P, Narrington N, Paustenbach D, Price P (1994) Recommended distribution for exposure factors frequency used in health risk assessment. *Risk Anal* 14:533-553
- Guo HR, Chiang HS, Hu H, Lipsitz SR, Monson RR (1994) Arsenic in drinking water and urinary cancers: a preliminary report. In: Chappell WR, Abernathy CO, Cothorn CR (eds) *Arsenic exposure and health*. Northwood, UK: Science and technology letters, pp119-124
- Hall LW Jr, Burton DT (1982) Effect of power plant coal pile and coal waste runoff and leachate on aquatic biota: an overview with research recommendations. CRC Press, Boca Raton, pp287-297
- Han BC, Jeng WL, Chen RY, Fang GY (1998) Estimation of target hazard quotients and potential health risk for metals by consumption of seafood in Taiwan. *Arch Environ Contam Toxicol* 35:711-720
- Han BC, Jeng WL, Hung TC, Wen MY (1996) Relationship between copper speciation in sediments and bioaccumulation by marine bivalves of Taiwan. *Environ Pollut* 91:35-39
- Han BC, Jeng WL, Jeng MS, Hung TC (1994) Copper intake and health threat by consuming seafood from copper contamination coastal environments in Taiwan. *Environ Toxicol Chem* 13:775-780
- Hsueh YM, Cheng GS, Wu MM, Yu HS, Kuo TL, Chen CJ (1995) Multiple risk factors associated with arsenic-induced skin cancer: effect of chronic liver disease and malnutritional status. *Br J Cancer* 71:109-114
- Jager T, den Hollander A, van der Poel P, Rikken GJ, Vermeire T (2001) Probabilistic environmental risk assessment for dibutylphthalate. *Human Ecol Risk Assess* 7:1681-1697
- Johnson RA, Gan DR, Berthouex PM (1995) Goodness-of-fit using very small but related samples with application to censored data estimation of PCB contamination. *Environmetrics* 6:341-348
- Liao CM, Chen BC, Singh S, Lin MC, Liu CW, Han BC (2002) Arsenic bioaccumulation and toxicity in tilapia (*Oreochromis mossambicus*) from blackfoot disease area in Taiwan. *Sci Total Environ*: submitted
- Lin MC, Liao CM, Liu CW, Singh S (2001) Bioaccumulation of arsenic in aquacultural large-scale mullet *Liza macrolepis* from blackfoot disease area in Taiwan. *Bull Environ Contam Toxicol* 67:91-97
- Reinert RE, Knuth BA, Kamrin MA, Stober QJ (1991) Risk assessment, risk management, and fish consumption advisories in the United States. *Fisheries* 16:5-12
- Singh S (2001) A physiologically based pharmacokinetic and pharmacodynamic model for arsenic accumulation in aquaculture fish from blackfoot disease area, Taiwan. Unpublished Ph.D. Dissertation, National Taiwan University, pp195
- Spehar RL, Fiandt JT, Anderson RL, Defor DL (1980) Comparative toxicity of arsenic compounds and their accumulation in invertebrates and fish. *Arch Environ Contam Toxicol* 9:53-63
- Suhendrayatna, Ohki A, Nakajima T, Maeda S (2002) Studies on the accumulation and transformation of arsenic in freshwater organisms. II. Accumulation and transformation of arsenic compounds by *Tilapia mossambica*. *Chemosphere* 46:325-331
- Sokal RR, Rohlf FJ (1995) *Biometry*. (3<sup>rd</sup> eds) W. H. Freeman and Company, New York, NY, USA
- Schoof RA, Eickhoff J, Yost LJ, Crecelius EA, Cragin DW (1999) Dietary exposure to inorganic arsenic. In: Chappell WR, Abernathy CO, Calderon RL (eds) *Arsenic exposure and health effects*. Elsevier, Oxford, UK, pp81-88
- Thompson RE, Voit EO, Scott GI (2000) Statistical modeling of sediment and oyster PAH contamination data collected at a south Carolina estuary (complete and left-censored samples). *Environmetrics* 11:99-119
- US EPA (US Environmental Protection Agency) (1989a) Guidance manual for assessing human health risks from chemically contaminated, fish and shellfish. EPA-503/8-89-002, US EPA Office of Marine and Estuarine Protection, Washington, DC
- US EPA (US Environmental Protection Agency) (1989b) Risk assessment guidance for superfund, human health evaluation manual, EPA-540/1-89-0002, US EPA Office of Emergency and Remedial Response, Washington, DC
- US EPA (US Environmental Protection Agency) (1997) Exposure factors handbook, general factors, EPA-600/P-95-002Fa, US EPA Office of Research and Development, Washington, DC
- US EPA (US Environmental Protection Agency) (2001) Integrated Risk Information System Database (IRIS), <http://www.epa.gov/iris>
- US FDA (US Food and Drug Administration) (1993) Guidance document for arsenic in shellfish. US Food and Drug Administration, Washington, DC, pp25-27
- Vermeire T, Jager T, Janssen G, Bos P, Pieters M (2001) A probabilistic human risk assessment for environmental exposure to dibutylphthalate. *Human Ecol Risk Assess* 7:1663-1679
- Vose D (2000) *Risk Analysis: A Quantitative Guide*. John Wiley & Sons, New York, NY, USA
- Zar JH (1999) *Biostatistical Analysis*. (4<sup>th</sup> ed) Prentice-Hall, Inc., New Jersey, USA