



# Model-based risk assessment for milkfish and tilapia exposed to arsenic in a traditional polyculture system with seasonal variations



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## ARTICLE INFO

### Article history:

Received 1 September 2013

Accepted 1 July 2014

### Keywords:

Milkfish

Tilapia

Arsenic

Bioaccumulation

Polyculture

Risk assessment

## ABSTRACT

For many decades, the traditional polyculture systems have contributed to increasing productivity throughout the Taiwan regions. It has been recognized that arsenic (As)-contaminated groundwater used for aquaculture in the southwestern coastal region of Taiwan is likely to pose a health threat to fish and humans. The purpose of this study was to assess farmed milkfish and tilapia exposure risk to As in a polyculture system using a model-based risk assessment framework. A first-order three-compartment model was used to simulate arsenic accumulation in fish and sediment appraised with the field-observed data. We constructed dose–response profiles obtained from acute toxicity bioassays to assess milkfish and tilapia exposure risk during different growing seasons. A probabilistic risk model was used to estimate the potential exposure risk. We showed that As accumulations in milkfish and tilapia were higher in summer than in fall. We found that there was a 20% probability for milkfish exceeding ~2% mortality in summer. However, waterborne As is not likely to pose a mortality risk for tilapia. Our results also revealed that tilapia benefited milkfish by reducing As concentrations in the water, indicating that tilapia can be biocontrol agents in the milkfish–tilapia polyculture systems. We suggest that the present mechanistic assessment framework can be used to assess exposure risk to environmental pollutants in polyculture systems and provide an appropriate exposure analysis to improve decision-making in the design and development of effective aquaculture systems.

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## 1. Introduction

Arsenic (As) in environments and ecosystems is an issue of concern. Arsenic and its compounds are mobile in the environment. A growing evidence indicated that the toxicity resulting from As exposure is connected to disturbed antioxidant homeostasis for aquatic organisms, such as increasing cytotoxicity, inducing oxidative stress, decreasing antioxidant enzyme activities, and altering the antioxidant system (Bagnyukova et al., 2007; Bhattacharya and Bhattacharya, 2007; Seok et al., 2007; Ventura-Lima et al., 2009).

In Taiwan, arsenic in groundwater may come from natural sources, such as de-watering of deep crustal fluids, seawater intrusion, and biogeochemical cycling of Fe, As, and S in alluvial sediments (Nath et al., 2008). Moreover, the over-pumping also introduces excess dissolved oxygen that may oxidize the mineral,

releases arsenic and increases the arsenic concentration in water (Liu et al., 2003).

Milkfish (*Chanos chanos*) and tilapia (*Oreochromis mossambicus*) are two commercially important species of fish stocked traditionally in Taiwan. Martinez et al. (2006) indicated that the production of milkfish is 8000–12,000 kg ha<sup>-1</sup> y<sup>-1</sup> for semi-intensive culture in Taiwan. On the other hand, tilapia production was nearly 70,000 metric tons per year and ranked fifth in the major seafood species in recent years (Taiwan Fisheries Agency, 2010). In Taiwan, most of the As-affected areas correspond to southwestern coastal regions. Unfortunately, aquaculture farms are mainly located in the southwestern coastal areas where As-contaminated groundwater is the predominant source of cultural water (Liao et al., 2003; Liu et al., 2006). Fish and shellfish have high potentials to accumulate metals via waterborne and/or dietary exposure pathways (Metian et al., 2008).

Polyculture is the simultaneously conducted culture of more than one species of fish with different feeding habits for an efficient increase of fish production (Cruz and Laudencia, 1980). In contrast to the monoculture, polyculture is potentially more sustainable

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for increased productivity through the reutilization of natural food resources and waste products of one species by another, and therefore the synergistic interactions improve when stocking population of different species reaches balance in the polyculture systems (Nunes et al., 2003; Langdon et al., 2004; Rahman et al., 2010; Davidson et al., 2013).

Ibrahim and Naggar (2010) found that the net profit of Nile tilapia (*Oreochromis niloticus*) and African catfish (*Clarias gariepinus*) with a polyculture system was significantly higher than monoculture. Cruz and Laudencia (1980) carried out an experiment on cultural milkfish with all-male Nile tilapia (*Tilapia nilotica*) and showed no effect on the growth and production of milkfish. A previous study reported that there was no food competition in the milkfish–tilapia polyculture system (MTPS) with a higher fish production than monoculture (IFP, 1976).

Tilapia feed selected large plankton, particularly zooplankton which results in a decrease in predatory pressure on a small phytoplankton and exerts high productivity. Aside from being a plankton feeder, tilapia cleans the pond bottom of detritus and decaying algae, and thus contributes in a way to keep good water quality (Vinyard et al., 1988). In nature, large schools of milkfish have been seen in near shore waters with well-developed reefs and in coastal lagoons. Milkfish are generally considered herbivorous. Lückstädt and Reiti (2002) indicated that the stomach contents of milkfish consist of a greater proportion of single-cell algae and less of diatoms, but also contained benthic and planktonic organisms and crustacean larvae. For the different food size selection and live area, it is not common for food competition among tilapia and milkfish.

In the traditional culture ponds, sediment is a common reservoir for many of the prevailing metals, such as Hg and As, containing elevated levels of potentially adverse metals in metal-contaminated sites (Wang et al., 2007; Chen et al., 2009). Generally, adsorption–desorption kinetics of metals exists in the sediment compartment (Clement and Faust, 1981; Monabbati, 1999; Hiller et al., 2007). Therefore, sediment in an aquaculture system plays a crucial role in determining water quality. Thus, to examine the bioaccumulation process of As in the MTPS, it is necessary to develop a complete dynamic model associated with major underlying factors affecting the As accumulation as it undergoes uptake/elimination and adsorption/desorption kinetics.

Milkfish has been polycultured with tilapia in Taiwan for many decades (ADAROC, 2014). Little is known, however, about risk assessment for groundwater As contamination of fish in the polyculture systems. In this study, we assess milkfish and tilapia exposure risks to waterborne As in the polyculture ponds using a model-based risk assessment framework.

To achieve this goal, we first constructed a MTPS model to predict the As burdens in milkfish and tilapia and validated these levels with field data. We then integrated the predict body burden and the dose–response relationships between internal lethal concentration and mortality obtained from the experimental studies to quantify As exposure risk in milkfish and tilapia. Moreover, fish exposure risks in different growing seasons were also included.

## 2. Materials and methods

### 2.1. Model structure

Waterborne As exposure is the major source for tilapia accumulation As. Huang et al. (2003) measured the As species in farmed tilapia and culture pond water and reported that the As accumulation in tilapia significantly increased with total As concentrations of pond water. Wang et al. (2007) analyzed the tilapia, water, and sediment samples that taken from 8 fishponds in the blackfoot disease areas and indicated that sediment was likely a sink of As in

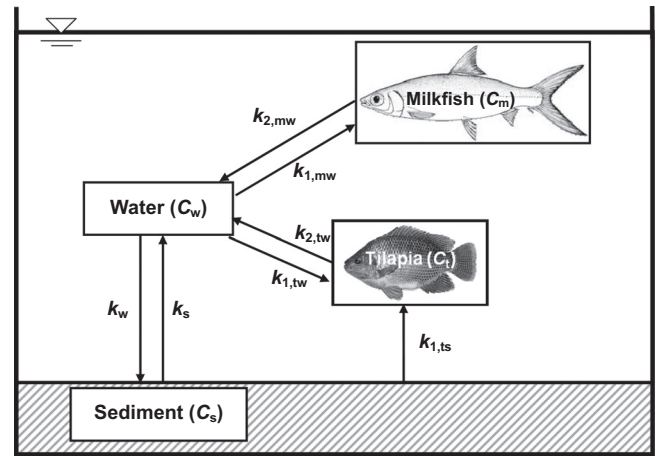


Fig. 1. Schematic showing the milkfish–tilapia polyculture system (MTPS) model that contains interactions among water, sediment, and fish compartments to describe As accumulations in milkfish *C. chanos*, tilapia *O. mossambicus*, and sediment (see Eqs. (1)–(3) for detailed definition).

the aquaculture ecosystem, and might have partially influenced the bioavailability of As to tilapia.

Thus, the fate and transport of As in a MTPS can be parsimoniously described by a first-order three-compartment model of milkfish–tilapia–sediment (Fig. 1). The scenario involves four major elements in a MTPS: (i) the amount of As in water is time-invariant and was regarded as the resource of As; (ii) the transport of As between compartments was modeled as a first-order process; (iii) As bioaccumulation in milkfish only considered waterborne exposure and the exposure via food was neglected, whereas As bioaccumulation in tilapia was based on uptake from both water and sediment due to the fact that tilapia favor the bottom layer as their habitat; and (iv) for both fish, decrease of body burden is through depuration from the whole body.

The first-order three-compartment model describing the concentrations of As in milkfish, tilapia, and sediment, respectively, and can be expressed as (Fig. 1),

$$\frac{dC_m}{dt} = k_{1,mw}C_w - k_{2,mw}C_m, \quad (1)$$

$$\frac{dC_t}{dt} = k_{1,tw}C_w + k_{1,ts}C_s - k_{2,tw}C_t, \quad (2)$$

$$\frac{dC_s}{dt} = k_wC_w - (k_{1,ts} + k_s)C_s, \quad (3)$$

where  $C_w$  is the As concentration of water ( $\mu\text{g L}^{-1}$ ),  $C_s$ ,  $C_m$ , and  $C_t$  are the time-varying As concentrations in sediment ( $\mu\text{g g}^{-1}$  wet wt), milkfish ( $\mu\text{g g}^{-1}$  wet wt), and tilapia ( $\mu\text{g g}^{-1}$  wet wt), respectively,  $t$  is the time of exposure (d),  $k_{1,mw}$  is the uptake rate constant from water in milkfish ( $\text{mL g}^{-1} \text{d}^{-1}$ ),  $k_{2,mw}$  is the elimination rate constant for As in milkfish ( $\text{d}^{-1}$ ),  $k_{1,tw}$  is the uptake rate constant from water in tilapia ( $\text{mL g}^{-1} \text{d}^{-1}$ ),  $k_{1,ts}$  is the uptake rate constant from sediment in tilapia ( $\text{g g}^{-1} \text{d}^{-1}$ ),  $k_{2,mw}$  is the elimination rate constant for As in tilapia ( $\text{d}^{-1}$ ),  $k_w$  is the coefficient of sedimentation and diffusion from water to sediment ( $\text{d}^{-1}$ ), and  $k_s$  is the coefficient of resuspension and diffusion from sediment to water ( $\text{d}^{-1}$ ).

We solved Eq. (1) to obtain the time-dependent As in milkfish ( $C_m(t)$ ) as

$$C_m(t) = C_{m,0}e^{-k_{2,mw}t} + \frac{k_{1,mw}}{k_{2,mw}}C_w(1 - e^{-k_{2,mw}t}), \quad (4)$$

where  $C_{m,0}$  is the initial As concentration at  $t=0$  ( $\mu\text{g g}^{-1}$  wet wt) in milkfish.

As concentration in sediment at the steady-state condition could be obtained by solving Eq. (3),

$$C_s = \frac{k_w}{k_{1,ts} + k_s} C_w. \quad (5)$$

We solved  $C_t(t)$  by substituting Eq. (5) into Eq. (2) as

$$C_t(t) = C_{t,0} e^{-k_2, tw t} + \frac{K}{k_{2, tw}} C_w (1 - e^{-k_2, tw t}), \quad (6)$$

where  $K = k_{1, tw} + (k_{1, ts} k_w / k_{1, ts} + k_s)$  and  $C_{t,0}$  is the initial As concentration at  $t = 0$  ( $\mu\text{g g}^{-1}$  wet wt) in tilapia.

Here, we defined the bioaccumulation factor (BCF) for milkfish as  $BCF_{mw} = k_{1, mw} / k_{2, mw}$  and  $BCF_{tw} = k_{1, tw} / k_{2, tw}$  for tilapia. On the other hand, the biota-sediment accumulation factor (BSAF) describing sediment-associated As into tissue of tilapia has the form as  $BSAF_{ts} = C_t / C_s$ .

## 2.2. Study data and model parameterization

We selected two wells in the Yun-Lin coastal area, a major arsenic-affected region, situated at the southwestern part of the alluvial fan of the Chou-Shui River in Taiwan as our study site. Yun-Lin is an aquaculture-based county in which groundwater is the major source of cultural water. In addition, high As concentrations are usually detected in the southwestern coastal area in Taiwan. Kuo et al. (2004) reported that As concentrations in well water ranged from  $\sim 30$  to  $\sim 500 \mu\text{g L}^{-1}$  in Yun-Lin. In this study, we used wells 1 and 2 to represent two wells containing high ( $500 \mu\text{g L}^{-1}$ ) and low ( $30 \mu\text{g L}^{-1}$ ) As, respectively, with seasonal variability in the period 1992–1998.

Many input parameters were needed to complete the MTPS including toxicokinetic parameters for milkfish and tilapia (i.e.,  $k_{1, mw}$ ,  $k_{2, mw}$ ,  $k_{1, tw}$ ,  $k_{1, ts}$ , and  $k_{2, mw}$ ). Therefore, we adopted the toxicokinetic parameter estimates from recent literature to complete this model (Chou et al., 2006; Tsai and Liao, 2006a,b; Wang et al., 2007). The exchange rates of water and sediment,  $k_w$  and  $k_s$ , can be estimated from published data (Clement and Faust, 1981; Monabbati, 1999) with the first-order kinetic models  $dC_w/dt = -k_w C_w$  and  $dC_w/dt = k_s C_s (W_s/V_w)$ , respectively.

To validate the MTPS model, a blanket search was conducted to collect recent publications that focused on As accumulations in milkfish, tilapia, and sediment in Taiwan aquaculture (Huang et al., 2003; Chou et al., 2006; Wang et al., 2007; Lin and Liao, 2008). Then, the estimated distributions of waterborne As concentrations were incorporated into the MTPS model to estimate the As levels in milkfish, tilapia, and sediment.

## 2.3. Dose–response analysis

As-induced mortality data in milkfish and tilapia resulted from recent experimental studies (Liao et al., 2005; Chou et al., 2006) were adopted for examining survival effects of As exposures. In brief, milkfish were exposed to As concentrations ranging from 0 to  $1000 \text{ mg L}^{-1}$  As (Chou et al., 2006). Liao et al. (2005) carried out a mortality experiment for tilapia exposure to concentrations of 0, 2, 10, 30, 50, and  $100 \text{ mg L}^{-1}$  As. Based on the toxicity data, As burdens in fish can be calculated by multiplying the BCF of each species. The relationships between As burdens in fish and mortality were then constructed.

We reanalyzed the experimental data and reconstructed the dose–response relationships between As burden in fish and mortality by employing the Hill model as (Garnier-Laplace et al., 2006),

$$M(C) = \frac{1}{1 + (C/ILC50)^n}, \quad (7)$$

**Table 1**

Values of parameter used in the milkfish–tilapia polyculture system model.

Parameter	Value	References
Milkfish ( <i>C. chanos</i> )		
$k_{1, mw}$ ( $\text{mL g}^{-1} \text{d}^{-1}$ )	$0.31 \pm 0.08^a$	Chou et al. (2006)
$k_{2, mw}$ ( $\text{d}^{-1}$ )	$0.25 \pm 0.18$	Chou et al. (2006)
Tilapia ( <i>O. mossambicus</i> )		
$k_{1, tw}$ ( $\text{mL g}^{-1} \text{d}^{-1}$ )	$0.481 \pm 0.072$	Tsai and Liao (2006b)
$k_{2, tw}$ ( $\text{d}^{-1}$ )	$0.164 \pm 0.063$	Tsai and Liao (2006b)
$k_{1, ts}$ ( $\text{g g}^{-1} \text{d}^{-1}$ )	$3.9 \times 10^{-3} \pm 1.3 \times 10^{-3}$	Wang et al. (2007)
Water–sediment		
$k_w$ ( $\text{d}^{-1}$ )	$5.28 \times 10^{-2}$	Monabbati (1999)
$k_s$ ( $\text{d}^{-1}$ )	$6.9 \times 10^{-4}$	Clement and Faust (1981)

<sup>a</sup> Mean  $\pm$  SE.

where  $C$  is the As concentration in fish ( $\mu\text{g g}^{-1}$  wet wt),  $M(C)$  is the  $C$ -dependent response measured as mortality (%), ILC50 is the internal effective concentration at 50% mortality, and  $n$  is the Hill coefficient.

## 2.4. Risk assessment model

In this study, we used the dose–response profiles describing the relationships between As level in fish and mortality as the conditional probability of  $P(M_m|C_m)$  and  $P(M_t|C_t)$  for milkfish and tilapia, respectively. As burdens in milkfish ( $C_m$ ) and tilapia ( $C_t$ ) were estimated by Eqs. (4) and (6) based on waterborne As concentrations in fish growing seasons. Exposure risks of milkfish and tilapia to As can be calculated by multiplying probability density function (PDF) of  $C_m$  and  $C_t$  and conditional probabilities of mortality ( $P(M_m|C_m)$  and  $P(M_t|C_t)$ ), respectively.

Therefore, a joint probability function (JPF) can be used to calculate the milkfish/tilapia risk probabilities,

$$R(M_m) = P(C_m) \times P(M_m|C_m), \quad (8)$$

$$R(M_t) = P(C_t) \times P(M_t|C_t), \quad (9)$$

where  $R(M_m)$  and  $R(M_t)$  represent the milkfish and tilapia mortality risk estimates based on As burdens in milkfish ( $C_m$ ) and tilapia ( $C_t$ ), respectively.

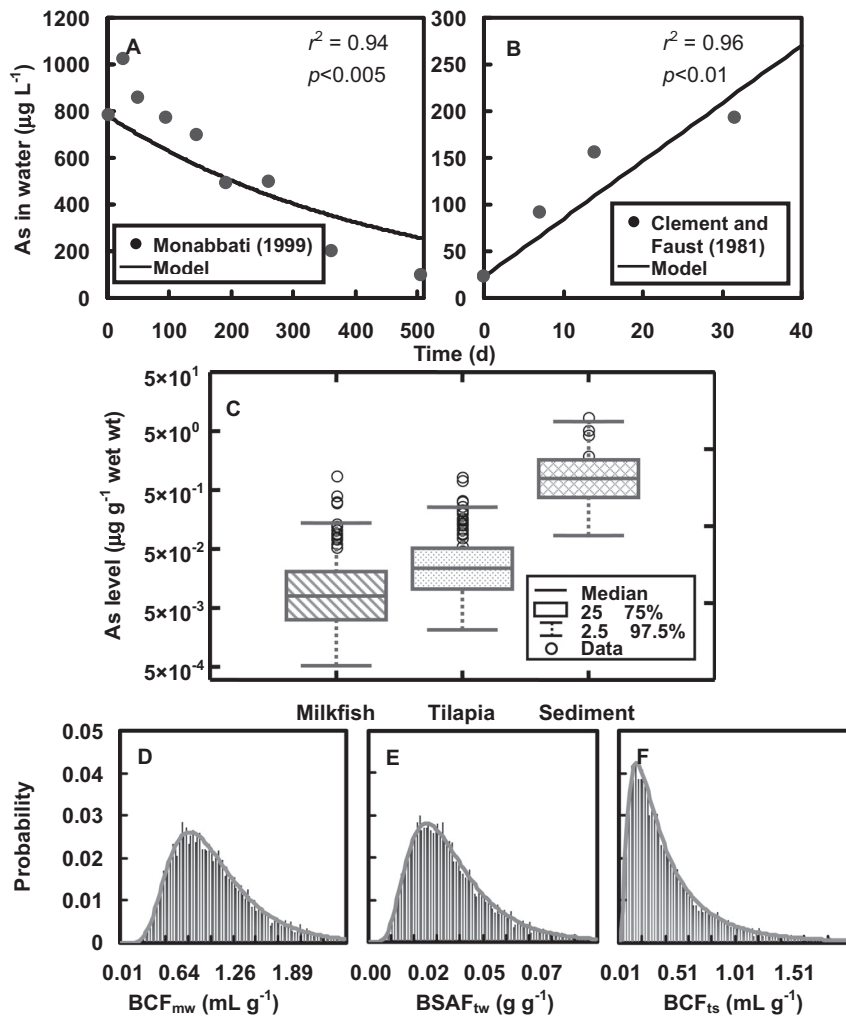
## 2.5. Uncertainty and data analyses

TableCurve 2D (Version 5.0, AISN Software Inc., Mapleton, OR, USA) and Statistica® software (Version 6.0, StatSoft, Tulsa, OK, USA) were used to optimally perform the model fittings. A value of  $p < 0.05$  was judged significant. We generated 2.5th and 97.5th percentiles as the 95% confidence interval (CI) for all fitted models and explicitly quantified the uncertainty of data by implementing a Monte Carlo technique (Crystal Ball® software Version 2000.2, Decisioneering, Inc., Denver, Colorado, USA). The results showed that 10,000 iterations were sufficient to ensure the stability of results. The MTPS model simulation was performed by Berkeley Madonna: Modeling and Analysis of Dynamic Systems (Version 8.3.9, <http://www.berkeleymadonna.com>).

## 3. Results

### 3.1. As levels in fish and sediment

The essential toxicokinetic parameter estimates for milkfish and tilapia were listed in Table 1. A first-order kinetic model was used to optimally fit the published data to obtain the exchange rates between water and sediment of  $k_w$  and  $k_s$  (Table 1; Fig. 2A and B). Our results showed that a lognormal (LN) probability model can



**Fig. 2.** Estimates for the exchange rates of water and sediment, (A)  $k_w$  and (B)  $k_s$ , calculated from published data with a first-order kinetic model. (C) Box and whisker plot representation of predicted distributions of As concentrations in milkfish, tilapia, and sediment estimating subjected to waterborne As concentrations from references to compare the predicted values with actual field data. Results of probabilistic distributions of (D)  $\text{BCF}_{mw}$ , (E)  $\text{BSAF}_{tw}$ , and (F)  $\text{BCF}_{ts}$  were determined by a lognormal model.

best-fit the published data of As concentrations in Taiwan aquaculture farms, resulting in a geometric mean (gm) of  $24.83 \mu\text{g L}^{-1}$  with a geometric standard deviation (gsd) of 3.08 ( $\text{LN}(24.83 \mu\text{g L}^{-1}, 3.08)$ ).

Based on the estimated As concentration distribution, the As levels in milkfish, tilapia, and sediment were then predicted by the MTPS model (Fig. 2C). Compared with the field data, our results showed that all the field data of As levels in milkfish, tilapia, and sediment were higher than the 75th-percentile predictions (Fig. 2C). However, most of the field data still fell within the prediction range of 2.5th- and 97.5th-percentiles (Fig. 2C). Fig. 2D–F illustrates the best-fit probability distributions for  $\text{BCF}_{mw}$ ,  $\text{BCF}_{tw}$ , and  $\text{BSAF}_{ts}$  with  $\text{LN}(0.32 \text{ mL g}^{-1} \text{ wet wt}, 2.39)$ ,  $\text{LN}(0.95 \text{ mL g}^{-1} \text{ wet wt}, 1.57)$ , and  $\text{LN}(0.03 \text{ g g}^{-1} \text{ wet wt}, 1.65)$ , respectively.

### 3.2. Exposure assessment

Based on the original measurements of well water with seasonal variation in Yun-Lin in the period 1992–1998 (Fig. 3A and C), the probability of distributions of As concentrations in wells 1 and 2 were obtained (Fig. 3B and D). The resulting distributions of As concentrations were  $\text{LN}(472.72 \mu\text{g L}^{-1}, 1.58)$  in summer and  $\text{LN}(390.26 \mu\text{g L}^{-1}, 1.59)$  in fall for well 1, whereas well 2 had As concentration distributions with  $\text{LN}(26.46 \mu\text{g L}^{-1}, 1.57)$  in summer and  $\text{LN}(20.37 \mu\text{g L}^{-1}, 1.69)$  in fall (Fig. 3B and D).

### 3.3. Dose–response model

The relationship between As body burden and mortality for milkfish can be well described with a Hill-based dose–response profile with a fitted Hill coefficient  $n = 2.05 \pm 0.47$  (mean  $\pm$  SE) and an effective As concentration in tissue at 50% mortality  $\text{LC}_{50} = 2.30$  (95% CI: 1.58–3.03)  $\mu\text{g g}^{-1}$  wet wt ( $r^2 = 0.95$ ,  $p < 0.05$ ) (Fig. 4A). For tilapia, the Hill model can also well depict the association between As body burden and mortality with  $n = 5.72 \pm 2.50$  and  $\text{ILC}_{50} = 30.45$  (95% CI: 24.51–36.39)  $\mu\text{g g}^{-1}$  wet wt ( $r^2 = 0.97$ ,  $p < 0.05$ ) (Fig. 4B). In view of the  $\text{ILC}_{50}$  estimates, tilapia were less sensitive to As than milkfish.

### 3.4. Risk estimates

Our results indicated that As body burdens in milkfish at wells 1 and 2 were 0.15 and  $0.009 \mu\text{g g}^{-1}$  wet wt, respectively, in summer and higher than those in fall of 0.13 and  $0.007 \mu\text{g g}^{-1}$  wet wt at wells 1 and 2, respectively (Fig. 5A and D). On the other hand, As levels in tilapia in summer were 0.45 and  $0.025 \mu\text{g g}^{-1}$  wet wt at wells 1 and 2, respectively, and were also higher than those in fall of 0.37 and  $0.019 \mu\text{g g}^{-1}$  wet wt at wells 1 and 2, respectively (Fig. 5G and J).

Table 2 summarizes the estimated milkfish and tilapia exposure risks for mortality at 20, 50, and 80% probabilities subject to

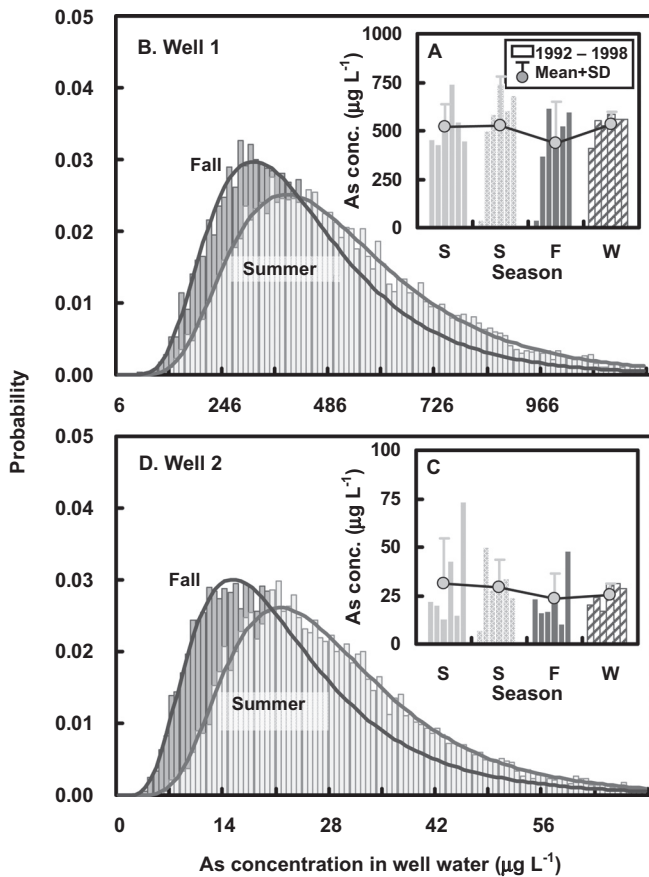


**Table 2**

Milkfish and tilapia exposure exceedance risk estimates for mortality (%) at 20, 50, and 80% probabilities subject to waterborne As concentration in growing seasons of summer and fall in study sites of well 1 (high level) and well 2 (low level) in Yun-Lin.

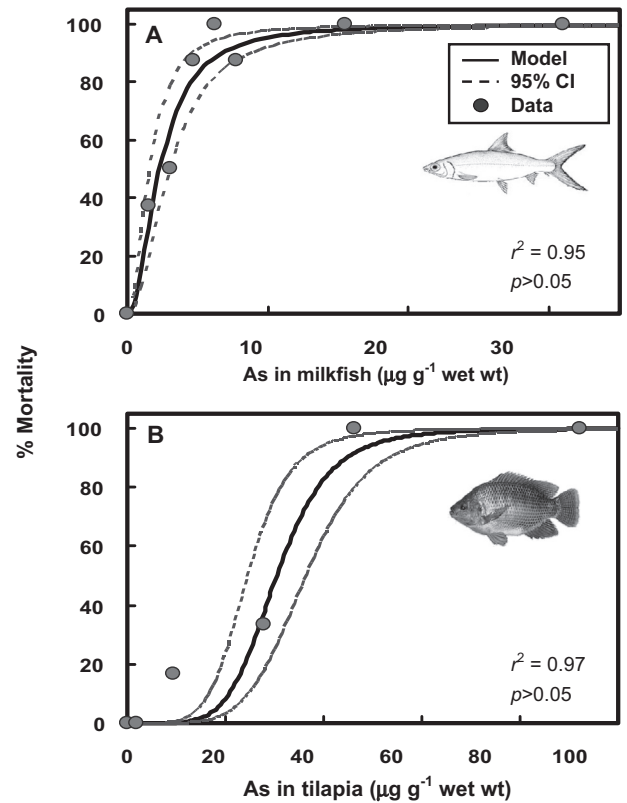
Species	Risk probability		
	20%	50%	80%
<b>Well 1</b>			
Milkfish			
Summer	1.77 (1.02–3.74) <sup>a</sup>	$3.61 \times 10^{-1}$ ( $2.07 \times 10^{-1}$ – $7.76 \times 10^{-1}$ )	$5.74 \times 10^{-2}$ ( $3.29 \times 10^{-2}$ – $1.24 \times 10^{-1}$ )
Fall	1.18 ( $6.77 \times 10^{-1}$ –2.51)	$2.39 \times 10^{-1}$ ( $1.37 \times 10^{-1}$ – $5.15 \times 10^{-1}$ )	$3.81 \times 10^{-2}$ ( $2.18 \times 10^{-2}$ – $8.21 \times 10^{-2}$ )
Tilapia			
Summer	$6.04 \times 10^{-8}$ ( $2.18 \times 10^{-8}$ – $2.09 \times 10^{-7}$ )	$2.59 \times 10^{-9}$ ( $9.34 \times 10^{-10}$ – $8.95 \times 10^{-9}$ )	$1.29 \times 10^{-10}$ ( $4.65 \times 10^{-11}$ – $4.46 \times 10^{-10}$ )
Fall	$1.65 \times 10^{-8}$ ( $5.94 \times 10^{-9}$ – $5.69 \times 10^{-8}$ )	$8.44 \times 10^{-10}$ ( $3.05 \times 10^{-10}$ – $2.92 \times 10^{-9}$ )	$3.55 \times 10^{-11}$ ( $1.28 \times 10^{-11}$ – $1.23 \times 10^{-10}$ )
<b>Well 2</b>			
Milkfish			
Summer	$4.95 \times 10^{-3}$ ( $2.83 \times 10^{-3}$ – $1.07 \times 10^{-2}$ )	$9.31 \times 10^{-4}$ ( $5.33 \times 10^{-4}$ – $2.01 \times 10^{-3}$ )	$1.48 \times 10^{-4}$ ( $8.46 \times 10^{-5}$ – $3.19 \times 10^{-4}$ )
Fall	$2.97 \times 10^{-3}$ ( $1.70 \times 10^{-3}$ – $6.40 \times 10^{-3}$ )	$5.47 \times 10^{-4}$ ( $3.13 \times 10^{-4}$ – $1.18 \times 10^{-3}$ )	$1.02 \times 10^{-4}$ ( $5.82 \times 10^{-5}$ – $2.20 \times 10^{-4}$ )
Tilapia			
Summer	$3.35 \times 10^{-15}$ ( $1.21 \times 10^{-15}$ – $1.16 \times 10^{-14}$ )	$1.73 \times 10^{-16}$ ( $6.24 \times 10^{-17}$ – $5.98 \times 10^{-16}$ )	$8.05 \times 10^{-18}$ ( $2.91 \times 10^{-18}$ – $2.78 \times 10^{-17}$ )
Fall	$9.97 \times 10^{-16}$ ( $3.60 \times 10^{-16}$ – $3.45 \times 10^{-15}$ )	$4.43 \times 10^{-17}$ ( $1.60 \times 10^{-17}$ – $1.53 \times 10^{-16}$ )	$1.20 \times 10^{-18}$ ( $4.33 \times 10^{-19}$ – $4.15 \times 10^{-18}$ )

<sup>a</sup> Median (95% CI).



**Fig. 3.** Seasonal variation of waterborne As concentrations in the period 1992–1998 in spring (S), summer (S), fall (F), and winter (W) and optimized lognormal distributions of As concentration in growing seasons of summer and fall at (A, B) well 1 with a high As level and (C, D) well 2 with a low As level.

As exposure in wells 1 and 2. Not surprisingly, exposure risks for both milkfish and tilapia were relatively higher in summer than those in fall (Fig. 5B, C, E, F, H, I, K and L). In summer, there was 20% probability for milkfish mortality exceeding ~2% at well 1 and only  $\sim 5 \times 10^{-3}\%$  at well 2 (Table 2, Fig. 5B and E). In contrast to milkfish, tilapia experienced relative lower risk probabilities of mortality



**Fig. 4.** Reconstructed dose–response profiles for the relationships between As concentration in whole body and mortality for (A) milkfish and (B) tilapia.

ranging from  $6.04 \times 10^{-8}$  to  $3.55 \times 10^{-11}\%$  even when exposed to the high As-enriched well water (Table 2, Fig. 5H).

#### 4. Discussions

##### 4.1. Risk assessment in MTPS

In Taiwan, raising fish in traditional polyculture systems has been considered as an efficient way to produce fish while reducing some of the problems associated with aquaculture because of the complementary use of scarce land and water resources. The traditional MTPS in Taiwan is also important for the food it produces

(e.g., protein) and for its reduced use of chemical fertilizers and pesticides relative to intensive, high-input aquaculture. In this study, we performed a model-based risk assessment for As exposure in MTPS at As-affected areas where intensive groundwater extraction is causing land subsidence of up to  $14.3 \text{ cm y}^{-1}$  (Tung and Hu, 2012).

Our probabilistic risk assessment framework was applied to predict As bioaccumulations and mortality risk with seasonal variations in MTPS from As-enriched well water as the predominant source of cultural water in southwestern coastal regions of Taiwan. We found that in a MTPS (i) As accumulations in milkfish and tilapia were higher in summer than in fall, (ii) there was a 20% probability for milkfish mortality exceeding  $\sim 2\%$  in summer, (iii) waterborne As poses insignificant mortality risk to tilapia, and (iv) tilapia benefited milkfish by reducing As concentration, indicating that tilapia can be biocontrol agents in the MTPS.

A comparison of the model predictions to field data is difficult due to difference in exposure conditions and the modifications in model (Li et al., 2010). The main reason may be due partly to the relative low uptake rate constant that we adopted from the laboratory experiments. However, the higher bioaccumulation was found at the field-derived condition than that of a laboratory-derived condition even at closed exposure concentration (Wang et al., 2007; Toni et al., 2011). Therefore, the lower predictions of body burden were assessed via the relative lower uptake rate constant. Moreover, the fish feed could also contain As, yet the proposed model only considered waterborne exposure. In addition, the differences in the comparison may also be induced by modifications in the model in that the bioaccumulation of metals can also be affected by biological (size, age, sex, growth rate, and physiological conditions) and abiotic factors (salinity, pH, and hydrochemistry of water) (Tsai and Liao, 2006a; Irina and Ronny, 2009; Li et al., 2010; Burger and Gochfeld, 2011).

The BCF was used to estimate the metal bioaccumulation potential of aquatic organisms. Based on the toxicokinetic parameters, we estimated the BCF through As concentrations in water. It appears that the BCF in tilapia is higher than in milkfish, suggesting that tilapia had higher uptake capacity. The estimated BCF of tilapia was  $0.95 \text{ mL g}^{-1}$  wet wt (i.e., nearly  $3.8 \text{ mL g}^{-1}$  dry wt), which was in good agreement with that obtained from the laboratory experiments of  $3.21 \text{ mL g}^{-1}$  dry wt (Chen and Liao, 2004) and  $2.68 \text{ mL g}^{-1}$  dry wt (Tsai and Liao, 2006b). However, it was different from Wang et al. (2007) and Huang et al. (2003) in the field studies. Wang et al. (2007) measured the As concentrations of tilapia, pond water, and sediment in the aquaculture system, resulting in a BCF of tilapia of  $55.0 \pm 40.1 \text{ mL g}^{-1}$  dry wt. Similarly, the BCF was in a range of  $2.8\text{--}119.6 \text{ mL g}^{-1}$  dry wt from Huang et al. (2003). On the other hand, we found a BCF of  $1.28 \text{ mL g}^{-1}$  dry wt in milkfish that was lower than that in the field of  $4.25\text{--}14.09 \text{ mL g}^{-1}$  dry wt (Lin and Liao, 2008). In the field, fish can be exposed to a lower background concentration for a long duration, hence tilapia has a relatively higher accumulation in the field; furthermore, this study did not consider the dietary exposure so that may be the potential reason for underestimating predictions.

To obtain the time-course accumulation in sediment, the exchange rates of water and sediment were obtained and described by the first-order rate constants. However, the mobility and contents of As are affected by pH of the sediment, ionic strength (such as Fe, Mn, and Al oxyhydroxides) and As loading (Vithanage et al., 2006; Hiller et al., 2007). Although we did not consider these factors in the present modeling,  $k_s$  was estimated to be  $6.9 \times 10^{-4} \text{ d}^{-1}$ , a value that is consistent with the estimate by Hiller et al. (2007). Our results showed that sediment accumulated much higher amount of As. This result supports that the sediment is the major sink for trace metals and is likely to pose potential risk for aquatic organisms (Wang et al., 2007; Chen et al., 2009).

Currently, the consumption of tilapia from the aquaculture in Taiwan poses limited or no food safety standard with respect to As. Action levels, tolerances and guidance levels for poisonous or deleterious substances in seafood have been developed by the US FDA (2009). The action levels established by the FDA for As in crustacea and molluscan shellfish were 76 and 86 ppm, respectively (US FDA, 2009). Comparing with this study, the levels of As in the tilapia ( $0.019\text{--}0.45 \text{ ppm}$ ) and milkfish ( $0.007\text{--}0.15 \text{ ppm}$ ) were found to be well less than the safety action levels set forth by US FDA. However, the action levels of As were not based on fish data. Nowadays, little is known about the accumulation of metal in freshwater fish from cultural systems and more research is needed to develop standard guidelines.

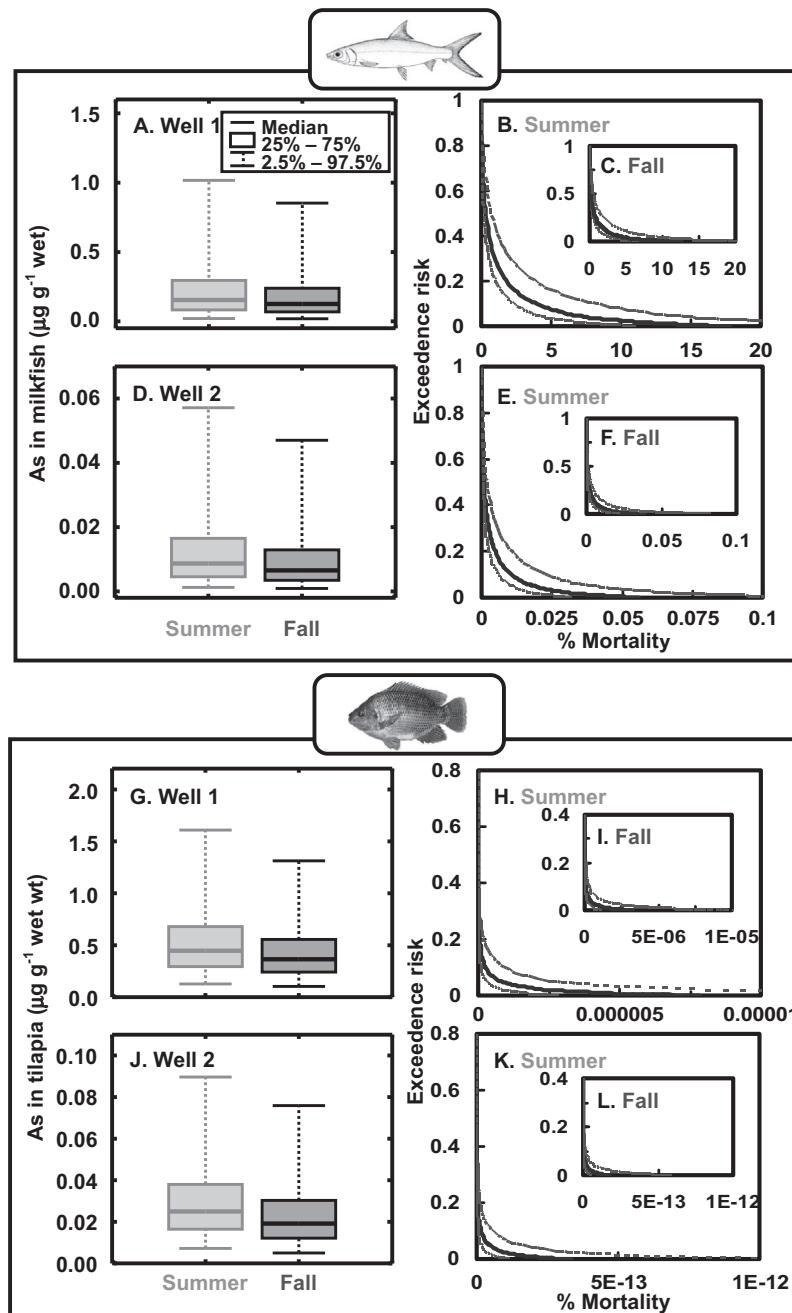
#### 4.2. Limitations and implications

We have demonstrated that the model performed well when validated with the field data. Our model also gives first estimates of As accumulation in MTPS. It is possible to apply the models in a predictive application through continued fundamental research and additional experience drawn from site-specific of well-characterized aquaculture systems.

As our knowledge, no published data have been available so far related to milkfish and tilapia exposed to As in the polyculture systems in Taiwan. In the present study, we attempted to incorporate the MTPS model with the field data related to milkfish and tilapia As accumulations in the monoculture system to assess the As exposure risk. However, due to the limited data on the polyculture systems in Taiwan and some essential data required for the modeling, the predicted risks associated uncertainties and variability would be increased. We recognize that our modeling cannot capture all factors that may affect the results of experiment. Therefore, this study performed the Monte Carlo simulation to obtain all likely effects. However, more polyculture systems data in Taiwan are needed to validate the estimated risks induced by As exposures in the future work.

However, no model can incorporate all the complexities of nature or make extra predictions of outcomes. The growth dilution and transformation of the As by metabolism of fish were negligible. Besides, the quality of hydrochemistry data was not considered in this study, although it may have an impact on results as well. Due to the limitation in the data of dose–response relationships, we can only find the endpoints of acute toxicity (mortality). However, the fact that our As exposures are relatively lower than adopted dose–response profiles, it may increase the uncertainty of risk assessment. If enough data are available to develop low concentration dose–response relationships, it will provide more reliable risk assessments than using the endpoint of mortality.

Many studies have indicated that seasonal variation affected the bioaccumulations of metals in fish (Ersoy and Çelik, 2010; Saei-Dehkordi et al., 2010). Metal bioaccumulation could be influenced by the season in association with many factors such as growth and reproductive cycle and the temperature and the pH of water (Ersoy and Çelik, 2009). In this study, we only used the waterborne As concentrations in summer and fall as inputs to the MTPS model on the basis that both seasons are growing seasons for milkfish ([http://163.26.66.5/teach/country/new\\_page\\_69.htm](http://163.26.66.5/teach/country/new_page_69.htm)). Arsenic concentrations were higher in summer than in fall, which may be influenced by water temperature (Köck et al., 1996). Tekin-Özan (2008) implied that metal concentrations increase in hot seasons and decrease in warm seasons. This study supports that the fall period can be recommended as the more appropriate season for catching fish in Yun-Lin aquacultures. In addition, the traditional risk assessments use the average environmental concentration for the exposure analysis. However, the growing seasons of fish are only in a specific season. Hence, careful consideration must be given



**Fig. 5.** Box and whisker plots showing the predicted As concentrations and estimated exceedance risk curves of As-induced mortality in summer and fall at wells 1 and 2 in milkfish (A–F) and tilapia (G–L).

to the selection of an appropriate exposure analysis to ultimately improve decision-making in the design and development of effective aquaculture systems.

## 5. Conclusions

Our MTPS model was built from existing culture systems and the approach could be used to predict the As accumulations in milkfish, tilapia, and sediment if particular environmental As concentrations are known, and vice versa. This present model is simple enough to be used in the aquaculture management context while addressing the complexity of a specific system. Most importantly, our study implicates mutually beneficial relationships between milkfish and tilapia in a polyculture system that undergoes environmental As pollution. We anticipate that this positive interaction and

complementary use of resources between milkfish and tilapia exposed to As-enriched cultural water can generate a biocontrol measure for improving aquaculture water quality and consequently increasing the productivity of traditional polyculture system. Furthermore, the dietary exposure is regarded as one of the main pathways of exposure of metal to the organisms (Ciardullo et al., 2008). For the future research, the dietary exposure can be incorporated into the MTPS model to predict As accumulation of fish in the polyculture systems. Moreover, it is very expensive to do pollution reduction at the source. We suggest that tilapia would be an additional means to reduce the pollution in the MTPS.

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