

Assessing carcinogenic risks associated with ingesting arsenic in farmed smeltfish (Ayu, Plecoglossus altirelis) in aseniasis-endemic area of Taiwan

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ABSTRACT

This study spatially analyzed potential carcinogenic risks associated with ingesting arsenic (As) contents in aquacultural smeltfish (Plecoglossus altirelis) from the Lanyang Plain of northeastern Taiwan. Sequential indicator simulation (SIS) was adopted to reproduce As exposure distributions in groundwater based on their three-dimensional variability. A target cancer risk (TR) associated with ingesting As in aquacultural smeltfish was employed to evaluate the potential risk to human health. The probabilistic risk assessment determined by Monte Carlo simulation and SIS is used to propagate properly the uncertainty of parameters. Safe and hazardous aquacultural regions were mapped to elucidate the safety of groundwater use. The TRs determined from the risks at the 95th percentiles exceed one millionth, indicating that ingesting smeltfish that are farmed in the highly As-affected regions represents a potential cancer threat to human health. The 95th percentile of TRs is considered in formulating a strategy for the aquacultural use of groundwater in the preliminary stage.

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1. Introduction

Arsenic (As) has been extensively documented to be a major risk factor for blackfoot disease (BFD) (Chen et al., 1994). Blackfoot disease was once epidemic on the southwestern coast of Taiwan (Tseng, 1977). The residents had used artesian well water with a high As content for over 50 years. Large-scale investigations of the association between As complexes in well water and ageadjusted mortality from various diseases (Lai et al., 1994; Chen et al., 1995a) and cancers (Wu et al., 1989; Chen and Wang, 1990) yield mutually supporting findings. According to statistical data, 2758 people had suffered from BFD in Taiwan before 1997 (Taiwan DOH, http://www.doh.gov.tw/EN2006/index_EN.aspx). Most of these patients are concentrated in few townships in which are thus called BFD hyperendemic areas. Similarly, the groundwater of the Lanyang Plain located in YiLan County of northeastern Taiwan contains arsenic levels that exceed the current Taiwan Environmental Protection Administration (Taiwan EPA) limit of 10 μ g/L. The residents there used high-arsenic artesian well water for over 50 years, and the arsenic concentration in some well water is up to 600 μ g/L or higher (Chiou et al., 1997). Most residential wells are less than 40 m in depth (Chiou et al., 2001) and contain arsenite and arsenate, which represent 87% and 5.8% of the total arsenic content, respectively, (Chen et al., 1995b). Significant dose-dependent relationships between the arsenic concentration in well water and an increased risks of cerebrovascular disease, urinary cancer and other cancers (Chiou et al., 1997, 2001), and adverse pregnancy outcomes (Yang et al., 2003) warrant further investigation.

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Groundwater is used liberally as an alternative to surface water in the Lanyang Plain, where surface water resources are severely deficient because of the high demand for water in irrigation as well as domestic, aquacultural and industrial uses. Nowadays, most inhabitants in this area do not drink well water directly since much epidemiological evidence has demonstrated that As exposure is strongly related to the incidence of diseases and cancer. However, very large quantities of groundwater are used to farm fish and shrimp. Arsenic in groundwater indirectly enters the food-chain through various paths and bio-accumulates in humans. Lin and Chiang (2002) collected samples of smeltfish and shrimp from various cultural ponds in the Lanyang Plain and found that levels of arsenic in smeltfish and shrimp were 25.6 and 16.65 μ g/g dry wt., respectively. Smeltfish is an aquacultural product with high economic value in Taiwan. The yield of smeltfish in the Lanyang Plain is as high as 92% of total yield of smeltfish in Taiwan, and this proportion is increasing



Fig. 1-(a) Study area in northeastern Taiwan. (b) Hydrogeological profile along A-B sections in (a).

annually (Taiwan FACOA, 2005). Most smeltfish is consumed domestically and the amount consumed increases annually (Taiwan FACOA, 2005). Smeltfish is farmed in freshwater, including groundwater and spring water. However, groundwater has become the major water source of water for ponds because the water temperature is stable and it is extracted easily (Han, 2003).

Risk assessment is frequently employed to quantify potential threats to human health through an exposurebioaccumulation-ingestion pathway of toxic substances in aquatic organisms (Pohl et al., 2003; Hung et al., 2004). Many studies have adopted the risk assessment approach to determine that the great amount of fish farmed in the BFD hyperendemic area and ingested was a severe threat to human health (Han et al., 1998; Liu et al., 2005; Ling et al., 2005; Jang et al., 2006). The regulatory maximum concentration of As in farmed pond water is 50 μ g/L in Taiwan (Taiwan EPA, 1998). Moreover, spatial distributions of contaminated groundwater quality are very heterogeneous. Time and cost limit analysis of in-situ data from field investigation. Measured data contains the considerable uncertainty. Geostatistics are widely used to model the spatial variability and distribution of field data with the uncertainty. Indicator kriging (IK) is frequently adopted as a non-parametric geostatistical approach. Indicator kriging makes no assumption regarding the distributions of variables, and uses a binary transformation of data to make the predictor robust (Cressie, 1993). In an unsampled location, the values estimated using IK represent the probability that does not exceed a particular threshold. Therefore, the expected value derived from the indicator data is equivalent to the cumulative distribution function of the variable (Smith et al., 1993). Indicator kriging has been frequently applied to estimate the pollution of soil by heavy metals. For example, Juang and Lee (1998), Castrignanò et al. (2000) and van Meirvenne and Goovaerts (2001) adopted IK to estimate the probability distribution of heavy metal pollution in fields and to delineate hazardous areas. Liu et al. (2004) and Goovaerts et al. (2005) used IK to evaluate the As pollution in groundwater and mapped the As-polluted extents of aquifers. Saisana et al. (2004) used IK to classify the zones that were polluted with nitrogen dioxide in air, as determined by regulatory standards. Furthermore, an indicator-based simulation, sequential indicator simulation (SIS), is also used to

Table 1 – Statistics regard	ing measured	concentration	ls of
As in groundwater			

Statistics	As concentration (μ g/L)		
Average	110		
Median	40		
Standard deviation	247		
Minimum	0.5		
Maximum	1010		
Percentiles			
27th	20		
47th	30		
62th	40		
80th	50		
90th	100		



Fig. 2-Vertical distribution of measured As concentrations.

describe the probability distributions of pollutants (Juang et al., 2004; Jang et al., 2006).

The objective of this work is to analyze spatially potential carcinogenic risks associated with ingesting As in aquacultural smeltfish (Ayu, Plecoglossus altirelis) in the Lanyang Plain of northeastern Taiwan. Sequential indicator simulation was used to reproduce As exposure distributions in groundwater based on their three-dimensional (3D) variability. Monte Carlo (MC) simulation was adopted to propagate the uncertainty of parameters concerning As exposure and bioaccumulation pathways. The target cancer risks associated with ingesting As contents in smeltfish were mapped to evaluate the potential risk to human health. The probabilistic risk assessment can be conducted to formulate suitable strategies under various remedial stages.

2. Materials and methods

2.1. Study area

The Lanyang Plain is located in YiLan County in northeastern Taiwan (Fig. 1), which is the alluvial fan of the Lanyang River. The area is triangular, with the Pacific Ocean to the east, Snow Mountain to the northwest and the Central Mountain to the southwest. The area is about 400 km² and each side is around 30 km long (Fig. 1a). Groundwater flows from west to east. The western sections of the plain near the mountains comprise the main recharging area of the groundwater, and the natural recharge is the source of groundwater (Peng, 1995). Unconsolidated sediments that underlie the alluvial fan contain abundant groundwater and is of the Quaternary age, and is partitioned into proximal-fan, mid-fan, and distal-fan areas (Chen, 2000). According to the core composition at different depths in the 22 hydrogeological investigation stations in the Lanyang Plain (Taiwan Central Geological Survey, available on http://www.hydro.meacgs.gov.tw/Rock13.htm), the

hydrogeological setting of the Lanyang Plain is roughly divided into three aquifers (Fig. 1b) (Lee et al., in press). The Water Resource Agency (Taiwan WRA) has set up 40 wells within depths of 18.9 m to 233 m to monitor the levels and background quality of groundwater, which is sampled and analyzed annually (http://gweb.wra.gov.tw/wrweb/). Most investigation stations have 1–4 monitoring wells, which are located in aquifers.

Agriculture, such as conducted in paddy and upland fields, is a common land use in this region. Aquacultural water-use also represents a major proportion of groundwater use in the Don-Shan, Jiao-Si and the coastal regions of the Lanyang Plain. However, areas in which smeltfish are farmed are located in Shan-Shin, Don-Shan and Wu-Jie: most of them are in Don-Shan (Han, 2003).

2.2. Arsenic concentrations in groundwater

A comprehensive survey of groundwater quality, which analyzed 40 monitoring wells using 25 hydrochemical parameters, was performed in the Lanyang Plain in 2004. Several taskoriented surveys of groundwater quality, which only focused on hydrochemical parameters in specific highly polluted wells, were performed, and a stable temporal pattern was identified during 2000–2003. Thus, this study adopted groundwater quality data from the survey in 2004 to assess health risk. Table 1 presents statistics on measured As concentrations in the study area. The average As concentration was 110 µg/L, and the maximum was 1010 µg/L. Approximately 15% of the As concentrations in the surveys were below 10 µg/L, which is the regulatory standard for drinking water in Taiwan. Fig. 2 plots the vertical distribution of the measured As concentrations. High As-contaminated concentrations (>200 µg/L) in groundwater were found at depths of 126 m to 183 m below the ground, and only in three wells.

2.3. Arsenic bioaccumulation pathways

The As concentrations in fish are bioaccumulation which results from fishpond environments and fish food. Fish

Fig. 3 – Conceptual diagram of three searching pairs for determining experimental variograms.

cancer and their distributions	Table 2–Pa	rameters	that	are	associated	with	risk	of
	cancer and t	heir distri	butio	ns				

Parameters	Distributions	Data sources
BWa (kg) α β	N(60.9, 9.9) ^a N(0.49, 0.025) 0.05	Taiwan DOH (1997) Japan MEXT (2005) Ling and Ling (2007)
BCF	LN(158.1, 1.98) ^b	Ling and Liao (2007) Lin and Chiang (2002), Ling and Liao (2007)
IR _{wt} (g/day wet wt)	LN(0.096, 1.24) ^c	FACOA (2005) and MOI (2005)

^a $N(\mu_a,\sigma_a)$ denotes a normal distribution with an arithmetic average of μ_a and an arithmetic standard deviation of σ_a .

 $^{\rm b}~{\rm LN}(\mu_{\rm g},\sigma_{\rm g})$ denotes a log-normal distribution with a geometric

average of $\mu_{\rm g}$ and a geometric standard deviation of $\sigma_{\rm g}.$

^c Valid range from 0.07 to 0.12 g/day wet wt.

bioaccumulation from farmed ponds is called bio-concentration and that in fish food is called bio-magnification (McGeer et al., 2003). Artificial diets for cultivated fish do not contain As, so bio-magnification can be ignored. Therefore, the As in smeltfish is concentrated mainly from the fishpond and can be expressed as follows.

$$C_{ayu} = BCF \cdot C_{pond} \tag{1}$$

where C_{ayu} and C_{pond} are the As concentrations in smeltfish and pond water, respectively. The BCF is the bio-concentration factor. Arsenic concentrations in cultivated ponds (C_{pond}) are considered to be the As concentrations in groundwater because groundwater was the major water source of pond water.

2.4. Geostatistical approaches

2.4.1. Variogram analysis

A geostatistical method is based on the regionalized variable theory which states that variables have both random and spatial structures in an area. A variogram of the data has to be determined first. An experimental variogram is computed to quantify the spatial variability of variables. The experimental

Table 3 – Mean square errors of fitting variograms						
Threshold	Model type	Horizontal direction	Vertical direction			
1st	Spherical	0.057	0.035			
	Exponential	0.052	0.025			
	Gaussian	0.067	0.040			
2nd	Spherical	0.047	0.069			
	Exponential	0.042	0.059			
	Gaussian	0.055	0.061			
3rd	Spherical	0.044	0.111			
	Exponential	0.041	0.087			
	Gaussian	0.054	0.110			
4th	Spherical	0.020	0.139			
	Exponential	0.019	0.103			
	Gaussian	0.020	0.124			
5th	Spherical	0.016	0.067			
	Exponential	0.015	0.047			
	Gaussian	0.016	0.076			





Fig. 4-Experimental variograms of indicator variables of As concentrations.

variogram is fitted by a theoretical model, $\gamma(h)$, which can be a spherical, exponential or Gaussian model, and three parameters of the fitted model – the nugget effect, the sill and the range – are determined. Variograms can be computed in different directions to detect any anisotropy of the spatial variability. An anisotropic model generally includes geometric anisotropy and zonal anisotropy. The geometric anisotropy yields variograms which have the same structural shape and variability (sill+nugget) but a direction-dependent range for the spatial correlation. Meanwhile, the zonal anisotropy is defined by the variation of the sills with the direction (Deutsch, 2002).

In the study area, the spatial scale in the horizontal direction differs markedly from that in the vertical direction: the minimum distance between pairs of groundwater monitoring wells in the horizontal direction exceeds 2 km, whereas the largest distance between two measurements of hydrochemical parameter concentrations at same location in the vertical direction is less than 0.25 km. This work thus used a three-dimensional (3D) spatial pairing strategy for determining experimental variograms in wells with the same elevation at different stations (Type I); in wells with different elevations at different stations (Type II) and in different wells at the same station (Type III). Fig. 3 presents a conceptual diagram of the 3D spatial pairing strategy. The first two types - Types I and II whose maximum angles between pair vectors and the horizontal plane are under 7°, are defined as a horizontal and large-scale searching pair. The third type (Type III) is defined as a vertical and small-scale search pair. The zonal anisotropy is perpendicular to the geometric anisotropy in this study. Their variabilities are individually calculated for different types of pairs. Thus, the vertical variability does not contribute to the horizontal variability with geometric anisotropy. When the analysis of the zonal anisotropy and geometric anisotropy is performed at the 3D space, the variabilities at the three principal axes are independent. A nugget effect is only the common variability. Deutsch (2002) discussed in detail the 3D anisotropic variability that was expressed as the vertical zonal variability with a small scale and the horizontal geometric variability with a large scale.

2.4.2. Indicator kriging

Indicator kriging is a non-parametric geostatistical method for estimating the probability that an attribute value is no greater than a specific threshold, z_k , at a given location u (Goovaerts, 1997). In IK, the spatial variable, Z(u), is transformed into an indicator variable with a binary distribution, as follows.

$$I\left(\mathbf{u}; z_{k}\right) = \begin{cases} 1, & \text{if } Z(\mathbf{u}) \leq z_{k}, \quad k = 1, 2, \dots, m\\ 0, & \text{otherwise} \end{cases}$$
(2)

The expected value of $I(u; z_k)$, conditional on *n* surrounding data, can be expressed as,

$$E[I(u; z_k|(n))] = Prob\{Z(u) \le z_k|(n)\} = F(u; z_k|(n))$$
(3)

where $F(u; z_k|(n))$ is the conditional cumulative distribution function (ccdf) of $Z(u) \leq_{Zk}$. Indicator kriging is an estimation technique which is based on an estimator that is defined as,

$$I^{*}(u_{0}; z_{k}) = \sum_{j=1}^{n} \lambda_{j}(z_{k})I(u_{j}; z_{k})$$
(4)

where $I(u_j; z_k)$ represents the values of the indicator at the measured locations, u_j , j = 1, 2, ..., n, and λ_j is a weighting factor of I $(u_j; z_k)$ that is used in estimating $I^*(u_0; z_k)$. This work used the gamv (variogram analysis program for irregularly spaced data) and ik3d (indicator kringing program) codes in Geostatistical Software Library and User's Guide (GSLIB) (Deutsch and Journel, 1998) to perform the experimental variogram and IK, respectively.

2.4.3. Sequential indicator simulation (SIS)

Sequential indicator simulation is the most widely used technique of the non-Gaussian simulation. It includes all original data and part values that are simulated previously within a neighborhood. A sequential simulation approach requires the simulation of a prior distribution at each unsampled location (Juang et al., 2004). In SIS, the IK estimator is first used to model the prior ccdf at each unsampled location. A linear interpolation yields a continuous ccdf within each class of threshold values (z_{k-1} , z_k]. The continuous ccdf at the lower tail is extrapolated toward a zero using a negatively skewed power model with ω =2.5 (as recommended by Goovaerts (1997) and Deutsch and Journel (1998)).

$$[F(z)]_{\text{Pow}} = \left[\frac{z - z_{\min}}{z_k - z_{\min}}\right]^{2.5} \cdot F^*(z_1)$$
(5)

The continuous ccdf at the upper tail is extrapolated toward an infinite upper bound using a hyperbolic model

Table 4 – Parameters fitted to the exponential model for five thresholds of As indicator variable								
Threshold	Direction	Nugget effect (c₀)	Sill (c)	Range (a) (km)	Max. range (km)	Min. range (km)	Anisotropic ratio	Max. variability direction
1 st	V	0.07	0.16	0.14	-	-	-	
	Н	0.07	0.16	14	17	8	2.12	N75°E
2nd	V	0.05	0.2	0.12	-	-	-	
	Н	0.05	0.2	9	16	6	2.67	N45°E
3rd	V	0.08	0.24	0.13	-	-	-	
	Н	0.08	0.18	10	15	7	2.14	N45°E
4th	V	0.1	0.31	0.13	-	-	-	
	Н	0.1	0.07	7	15	5	3	N15°E
5th	V	0.04	0.35	0.11	-	-	-	
	Н	0.04	0.06	6	10	4.5	2.2	N15°E
H: horizontal;	V: vertical.							



Fig. 5 - Processes for determining joint distributions and for assessing risk.

with ω = 1.5 (as recommended by Goovaerts (1997) and Deutsch and Journel (1998)).

$$[F(z)]_{Hyp} = 1 - \frac{z_k^{1.5} \cdot \left[1 - F^*(z_k)\right]}{z^{1.5}} \tag{6}$$

where $F^*(z_k)$ is the sample *ccdf* at z_k . A maximum attribute value of As concentrations in groundwater was set to $1010 \,\mu g/L$, which is the maximum value reported historically in this area, avoiding the occurrence of unexpected results (such as a very large and irrational concentration that was estimated at the 95th percentile).

Multiple realizations can be yielded via various random paths. Each realization followed a random path which represents likely the spatial distribution of As concentrations in groundwater. Therefore, numerous realizations can be used to evaluate the variation and uncertainty of the As concentrations. This work used the gamv (semi-variogram analysis) and sisim (sequential indicator simulation program) codes in GSLIB (Deutsch and Journel, 1998) to perform the experimental variogram and SIS, respectively. The As realizations that were reproduced by SIS are a non-parametric distribution and reproduced 250 data at each cell.

2.5. Assessment of risk to human health associated with ingesting smeltfish

Inorganic As species, arsenite(III) and arsenate(V), are typically more toxic than organic As species, such as arsenobetaine,



Aquifer 3

Fig. 6-GIS maps of the TRs based on the 95th percentile risk at aquifers 1-3 in the Lanyang Plain.

arsenocholine, MMA (monomethylarsonic acid), DMA (dimethylasinic acid), arsenosugar, and arsenolipid (Mandal and Suzuki, 2002; Oremland and Stotlz, 2003). The organic As forms are considered practically as a lower toxicity or nontoxicity (Shiomi et al., 1996). The US EPA (1988) has recommended that the uptake of inorganic As by various seafood species be used to determine potential risks, such as the target cancer risk (TR), to human health. A method for estimating TR values is supported by the US EPA Region III Risk-Based Concentration Table (US EPA, 1988, 2001, 2006). The risk of carcinogenic effects of inorganic As is expressed as the excess the probability of contracting cancer over a lifetime of 70 years.

Pacific Ocean

Lanyang R.

10.SuAd



A model for estimating target cancer risks (lifetime cancer risks) via ingesting smeltfish is (US EPA, 2001),

$$TR_{t} = \frac{EFr \times EDtot \times IR_{t} \times C_{it} \times CPSo}{BWa \times ATc} \times 10^{-3}$$
(7)

where TR_t is the target cancer risk value (the incremental individual lifetime cancer risk) for daily ingestion of smeltfish; EFr is the exposure frequency (350 days/year); EDtot is the exposure duration (30 years); IR_t is the ingestion rate for the edible portion of smeltfsh (g/day wet wt); C_{it} is the inorganic As concentration in the edible portion of smeltfish (μ g/g wet wt); CPSo is the oral carcinogenic potency slope (risk per (mg/kg/day)) (1.5 (mg/kg/day)⁻¹); BWa is the body weight of a Taiwanese adult (kg), and ATc is the averaging time for carcinogens (25,550 days).

Eq. (7) can be rewritten using the aforementioned parameters, the As exposure and the bioaccumulation pathways (Jang et al., 2006).

$$TR_{t} = \frac{IR_{t} \times C_{it}}{BWa} \times 6.16 \times 10^{-4}$$

$$= \frac{(IR_{wt} \times \alpha) \times \left[\left(\frac{C_{well}}{1000} \times BCF \right) \times \beta \right]}{BWa} \times 6.16 \cdot 10^{-4}$$
(8)

where IR_{wt} is the ingestion rate of whole smeltfish (g/day wet wt); α is the edible ratio of smeltfish; C_{well} is the estimated As concentration in groundwater locates around farmed ponds (μ g/L), and β is the ratio of inorganic As contents to total As contents in smeltfish. The arithmetic average α was 0.49 and the arithmetic standard deviation was 0.025 (Japan MEXT, 2005); the β value was 0.05 (Ling and Liao, 2007). IR_{wt} is a geometric mean value of 0.096 (g/day wet wt.) with a geometric standard deviation of 1.24, which values were calculated from the Taiwanese population of age>4 years (Taiwan MOI, 2005) and the annual consumption quantity of smeltfish from 2001 to 2005 (Taiwan FACOA, 2005). The arithmetic average of BWa was 60.9 kg and the arithmetic standard deviation was 9.9 kg (Taiwan DOH, 1997). The geometric average of BCF was 158.1 and the geometric standard deviation was 1.98 (Ling and Liao, 2007). The distributions of the parameters were identified using a Kolmogorov-Smirnov (K-S) test. Robust data concerning the parameters were generated using the MC simulation based on their measured distributions and their uncertainty was properly accounted for (Goovaerts et al., 2001; US EPA, 2001). The @Risk (Version 4.5, Professional Edition, Palisade Crop., USA) software was used to analyze statistically the measured data and to carry out the MC simulation. Table 2

lists statistical distributions of the parameters and the sources of the data.

3. Results and discussion

3.1. Variogram analysis and realizations of As concentrations

The measured As concentrations at the 27th, 47th, 62th, 80th, and 90th percentiles, as shown in Table 1, were used as the five thresholds (z_k , k=1,2,...,5). A lag of 0.01 km was used to analyze the small-scale vertical variograms of indicator variables of As concentrations. The nugget effect of a nested variogram is mainly inferred from the vertical variogram, which typically has the excellent spatial resolution. In the analysis of fitting variograms, an exponential variogam model was selected with minimized mean square errors (Table 3) to yield the best fitting in the vertical variograms (Fig. 4). The fitted vertical parameters of range, nugget effect and sill are 0.11-0.14 km, 0.04-0.1 and 0.16-0.35 (Table 4), respectively. A lag of 2.3 km was then used to analyze the horizontal omnidirectional variograms of indicator variables of As concentrations. For the analysis of zonal anisotropy, the fitted model of the horizontal variograms should be consistent with the exponential model that was used in the vertical variograms (Fig. 4). The fitted horizontal parameter of ranges and sills are 6-14 km and 0.06-0.2 (Table 4), respectively. Notably, the identical sills of the vertical and horizontal variograms occur at the 1st and 2nd thresholds. The zonal anisotropic model is equal to the geometric anisotropic model for the thresholds; zonal anisotropic model is a 3D geometric anisotropic model. Since a geometric anisotropic model has the same structural shape and variability (Deutsch and Journel, 1998), the model fitted to the omnidirectional horizontal variograms was also adopted to analyze the geometric anisotropic variability in the horizontal direction. The determined directions of maximal variability for each threshold range from N15°E to N75°E. The anisotropic ratio (maximum range/minimum range) ranges from 2.12 to 2.67 (Table 4). Indicator kriging was employed to estimate the probability distribution of arsenic concentrations based on variogram models. The study area was horizontally discretized using a grid of 338 cells, with a spacing of 1 km. All fitting parameters in the models was followed the cross-validation procedure (Deutsch and Journel, 1998; Chilès and Delfiner, 1999).

3.2. Analysis of uncertainty in risk assessment

This work probabilistically considered four parameters – BCF, α , BWa and IR_{wt} – in risk assessment, except for As concentrations in groundwater. The parameters were followed either a normal distribution or a log-normal distribution which was a two-parameter distribution. Before an MC simulation was implemented, normal and log-normal distributions were determined using an arithmetic average with an arithmetic standard deviation and a geometric average with a geometric standard deviation, respectively.

The four parameters in risk assessment were reproduced individually 250 data using a MC simulation based on their observed distributions to propagate their uncertainty. A series of joint distributions that integrated the aforementioned parameters were carried out (Goovaerts et al., 2001; Jang et al., 2006). Fig. 5 shows in detail the procedure for combining the joint distributions. Parameters with the same distribution shape had the priority in the combination procedure. Joint distributions that integrate two parameters was first produced — the IR_{wt} and BCF, and the $(1/BW_a)$ and α . The joint distributions with 250 data were newly identified using a K-S test. An arithmetic average and an arithmetic standard deviation for the normal distributions; or a geometric average and a geometric standard deviation for the log-normal distribution were computed from the joint distributions of 62,500 data. The joint distribution of the IR_{wt} and BCF parameters was fitted in a log-normal distribution of LN(11.02, 2.12). The joint distribution of the (1/BW_a) and α parameters was a normal distribution with N(0.008, 0.001). To reduce the complicated computation, 250 data were yielded from the joint distributions using an MC simulation. A joint distribution of four parameters was then produced. The joint distributions of BCF, IR_{wt}, α and (1/BWa) were consistent with a log-normal distribution of LN(0.811, 1.851). The As concentrations in groundwater is a non-parametric distribution, and were combined in the last process of the joint distributions for each cells.

The β value of 0.05 and the constant 6.16×10^{-4} in Eq. (8) were finally applied to calculate TR_t. Traditionally, the 5th, 25th, 50th, 75th and 95th percentiles of risks are displayed in a box-and-whiskers plot to assess the likelihoods of exceeding the corresponding risk levels (US EPA, 2001; Liu et al., 2005; Ling et al., 2005). In this work, the five percentiles of TRs at each cell were determined to assess spatially health risks associated with different probabilities.

3.3. Carcinogenic risks associated with ingesting arsenic in aquacultural smeltfish

The geographic information system (GIS) was applied to map TRs at three various depths. Irregular variations present in the TR maps due to the highly heterogeneity of 250 As realizations that are reproduced by SIS. All TRs determined from the 5th, 25th 50th and most 75th percentiles were below one millionth. All TRs determined from the 75th and 95th percentiles exceed one millionth at the cells at the three aquifers. Fig. 6 shows cell-level GIS maps of the 95th percentiles of TRs of three aquifers in Lanyang Plain. Four levels of TR, 0-1×10⁻⁶ (safe), 1-2×10⁻⁶ (hazardous), $2-10 \times 10^{-6}$ (hazardous) and $10-15 \times 10^{-6}$ (hazardous), are displayed. At aquifer 1, 12% of TRs exceeded one millionth and the largest TR was 2.12×10^{-6} , and hazardous regions were distributed at YiLan and Yuan-Shan. At aquifer 2, 10% of TRs exceeded one millionth and the largest TR was 14.3×10^{-6} , and most hazardous regions were at Don-Shan and Su-Ao. At aquifer 3, 25% of TRs exceeded one millionth and the largest TR was 12.74×10^{-6} , and most of the hazardous regions were mainly distributed at Lou-Don, Wu-Jie, Don-Shan and Su-Ao. Fig. 7 shows cell-level GIS maps of the 75th percentiles of TRs of aquifers 2 and 3 in Lanyang Plain. At the 75th percentile, the TRs of only one and two cells exceeded one millionth - at aquifers 2 and 3, respectively – and the largest TR was 6.55×10^{-6} and 5.84×10^{-6} . However, all TRs determined from the 75th percentiles were below one millionth at aquifer 1.

The hazardous regions at the 95th percentile of risks increase with increasing depth. The 95th percentile of risk is

a strict standard and may not be easily implemented currently. Thus, this study suggests that the 75th percentile of health risks is adopted to assess the risks of cancer associated with ingestion of As in aquacultural smeltfish in establishing the preliminary remedial framework. The 95th percentile of health risks computed from extrapolating ccdfs using a hyperbolic model and quantifying the parameter uncertainty using a MC simulation is considered with the extreme caution that is appropriate to the risks associated with maximal exposure (Jang et al., 2006). The spatial risk pattern at the aquifer 1 overtly differs from those at the aquifers 2 and 3. The regions of high risks are in the southern coastal area. Arsenic concentrations in pond water have contributed markedly to farmed smeltfish via bioaccumulation, posing a potential risk to human health through the foodchain (Ling et al., 2005). The As concentrations in pond water were strongly related to those measured in groundwater neighboring the pond. Accordingly, the spatial variation of As concentrations in groundwater should be considered to assess risk. Thus, the 95th percentile of health risks can be adopted to assess a land-use plan for aquaculture.

Since the use of groundwater as fishpond water has several advantages, such as convenience of acquisition, low expense of withdrawal and stable temperature in pond water, Ascontaminated groundwater is still used inevitably in the Lanyang Plain. This study proposed a risk-based scheme that is associated with the dynamic management of Ascontaminated groundwater. Ingesting rate can increase with the yield of aquacultural smeltfish increased, posing a risk to human health. Currently cultivated ponds at Don-Shan can be relocated to other safe regions. If ponds cannot be relocated, shallow groundwater can be extracted as a safe water source.

4. Conclusion

This work spatially analyzed the potential risks of cancer associated with the ingestion of As in aquacultural smeltfish in the Lanyang Plain of northeastern Taiwan. A probabilistic risk assessment by the MC and SIS were used to quantify uncertainty of parameters and to formulate proper strategies under various remedial phases. The 95th percentile of risks should be considered in the land-use plan for aquaculture. This investigation focuses on the use of groundwater with various levels of As exposure which induces the potential risk of cancer associated with the consumption of aquacultural smeltfish. Since TRs values exceed one millionth, the use of groundwater in aquaculture should be reduced or the use of spring water considered. Groundwater is a major water source that meets the needs of aquaculture in the Lanyang Plain. Regarding the development of aquacultural smeltfish businesses, this work suggests that smeltfish aquaculture should be relocated to other safe regions, or shallow groundwater or spring water should be used.

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