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Spatial Analysis of Potential Carcinogenic Risks Associated with Ingesting Arsenic in Aquacultural Tilapia (*Oreochromis mossambicus*) in Blackfoot Disease Hyperendemic Areas

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This work analyzed spatially potential carcinogenic risks associated with ingesting arsenic (As) contents in aquacultural tilapia (*Oreochromis mossambicus*) in coastal regions of southwestern Taiwan, where the blackfoot disease prevails. Sequential indicator simulation (SIS) was used to reproduce As exposure distributions in groundwater based on their three-dimensional variability. A target cancer risk (TR) associated with ingesting As in aquacultural tilapia was calculated to evaluate the potential risk to human health. Owing to sparse measured data, Monte Carlo simulation and SIS properly accounted for the uncertainty of assessed parameters. The probabilistic risk assessment formulated suitable strategies under various remedial stages. Aquacultural regions with high risks were mapped to elucidate the safety of groundwater use at different depths. Many TRs determined from the risks at the 75th and 95th percentiles exceed one millionth in the regions, indicating that ingesting tilapia farmed in the highly As-polluted regions poses potential cancer threats to human health. The 75th percentile of TR is considered in formulating a remedial strategy for the aquacultural use of groundwater in the preliminary stage. Additionally, this study suggests reducing the use of groundwater in aquaculture or changing the depths from which groundwater is withdrawn in the areas with high risks of cancer.

Introduction

Arsenic (As) has been well-documented to be a major risk factor for blackfoot disease (BFD) (*1*). Blackfoot disease was once common on the southwestern coast of Taiwan (*2*). The residents used artesian well water with a high As content for over 50 years. Large-scale investigations on the association between As complexes in well water and age-adjusted mortality from diabetes (*3*), hypertension (*4*), and cancers of the nasal cavity, lung, liver, bladder, kidney, and prostate (*5*, *6*) yield mutually supporting findings. Patients with the BFD had a markedly increased incidence of cancer, following adjustment for cumulative As exposure.

Groundwater has been used abundantly as an alternative to surface water in the coastal region of southwestern Taiwan, where surface water resources are severely deficient because of the high demand for water for domestic, irrigated, aquacultural, and industrial uses. Nowadays, most inhabitants in this area do not drink well water directly since epidemiological evidence proves that the As exposure is strongly related to the incidence of diseases and cancer. However, very large quantities of groundwater are used to farm fish and shellfish. Arsenic in groundwater indirectly enters the food chain via various paths and bioaccumulates in humans. Han et al. (*7*) found the total As concentrations of 0.13−1.45 μg/g dry wt bioaccumulated in tilapia in the BFD areas of Taiwan. Liao and Ling (*8*) reported the total As concentrations of 26.3 ± 16 to 251.7 ± 12.2 μg/L in pond water and 0.94 ± 0.3 to 15.1 ± 8.2 μg/g dry wt in farmed fish in this region.

Spatial distributions of contaminated groundwater quality are commonly heterogeneous. However, only a small proportion of in situ data can be analyzed in a field investigation owing to time and cost constraints. Consequently, sparsely measured data contain considerable uncertainty. Geostatistics is widely used in modeling the spatial variability and distribution of field data with uncertainty. Indicator kriging (IK), which is a frequently employed nonparametric geostatistical method, makes no assumption on distributions of variables, and a 0−1 indicator transformation of data is used to make the predictor robust to outliers. At an unsampled location, the values estimated by IK represent the probability that does not exceed a particular threshold. Indicator kriging has been frequently applied to soil pollution by heavy metals. For example, Juang and Lee (*9*), Castrignanò et al. (*10*), and van Meirvenne and Goovaerts (*11*) adopted IK to estimate the probability distribution of heavy metal pollution in fields and to delineate hazardous areas. Liu et al. (*12*) used IK to evaluate the As exposure probability and contamination potential in the Yunlin aquifer for the standard of As in drinking water. Saisana et al. (*13*) used IK to delineate the zones polluted with nitrogen dioxide in air, as determined by regulatory standards. Furthermore, an indicator-based simulation, sequential indicator simulation (SIS), also is used to delineate probability distributions of pollutants (*14*, *15*). An interpolation technique, such as kriging, can be regarded as a tool to help with evaluating spatial risks to human health. Vieira et al. (*16*) presented a method for producing geographic data on disease risks based on residential history data from a population-based case-control study. Crude and adjusted odds ratios were estimated by adaptive rate stabilization and mapped using kriging as an interpolative method to examine the risk of lung cancer. Yu et al. (*17*) employed kriging with depth trends to construct a map of As concentrations and to estimate As concentrations in groundwater. They proposed a combined geostatistical and epidemiological framework for evaluating human health and potential remedies for As-containing drinking groundwater in Bangladesh.

The objective of this work is to analyze spatially potential carcinogenic risks associated with ingesting As in aquacultural tilapia (*Oreochromis mossambicus*) in the BFD hyperendemic areas of southwestern Taiwan. Sequential indicator simula-

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tion was used to reproduce As exposure distributions in groundwater based on their three-dimensional (3-D) variability. Monte Carlo (MC) simulation was used to propagate the uncertainty of parameters concerning As exposure and bioaccumulation pathways and tilapia experiments. Target cancer risks of ingesting As contents in tilapia were mapped to assess the potential risk to human health. The probabilistic risk assessment can be used to formulate suitable strategies under various remedial stages.

Theory

Variogram Analysis. A geostatistical method is based on the regionalized variable theory that states that variables have both random and spatial structures in an area. A variogram of the data should first be determined. An experimental variogram is computed to quantify the spatial variability of variables. The experimental 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. In the study area, the spatial scale in the horizontal direction differs greatly from that in the vertical direction. Section A in our Supporting Information depicts in detail the 3-D anisotropic variability.

Indicator Kriging. Indicator kriging is a nonparametric geostatistical method for estimating the probability that the attribute value is no greater than a specific threshold, z_k, at a given location u (*18*). In IK, the spatial variable, $Z(u)$, is transformed into an indicator variable with a binary distribution, as follows.

$$I(u; z_k) = \begin{cases} 1, \text{ if } Z(u) \le z_k, \ k = 1,2,.....m \\ 0, \text{ otherwise} \end{cases} \quad (1)$$

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

$$E[I(u; z_k|(n))] = \text{Prob}\{Z(u) \le z_k|(n)\} = F(u; z_k|(n)) \quad (2)$$

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

$$I^*(u_0; z_k) = \sum_{j=1}^{n} \lambda_j(z_k) I(u_j; z_k) \quad (3)$$

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 $I(u_j; z_k)$ used in estimating $I^*(u_0; z_k)$.

Sequential Indicator Simulation (SIS). Sequential indicator simulation is the most widely used non-Gaussian simulation technique. This includes all original data and part previously simulated values within a neighborhood. A sequential simulation approach requires simulation of a prior distribution at each unsample location (*15*). In SIS, the IK estimator is first used to model the prior ccdf at each unsample 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 $\omega = 2.5$. The continuous ccdf at the upper tail is extrapolated toward an infinite upper bound using a hyperbolic model with $\omega = 1.5$ (as recommended by Goovaerts (*18*) and Deutsch and Journel (*19*)). A maximum attribute value of As concentrations in groundwater was set to 2500 mg/L, which was a maximum value reported historically in this area (*1, 2, 4, 6*), avoiding the occurrence of unexpected results (that is, a very large and irrational concentration estimated at the 95th percentile).

Multiple realizations can be yielded via various random paths. Each realization following a random path represents a likely result of the spatial distribution of As concentrations in groundwater. Therefore, numerous realizations can evaluate the variation and uncertainty of the As concentrations. This work used the gamv and sisim codes in GSLIB (*19*) to perform the experimental variogram and SIS, respectively.

Materials and Methods

Study Area and Hydrogeology. The study area is located in the plane region of Yunlin, Chiayi, and Tainan counties (Figure 1a). This area is enclosed by the Taiwan Strait to the west, the Central Mountain to the east, the Choushui river to the north, and the Erhjin river to the south. The northern study area, Yunlin County, is the southern part of the Choushui River alluvial fan, and the central and southern study area, Chiayi and Tainan counties, covers the Chianan plain. The region is approximately 2700 km^2 and extends 30 km from east to west and 90 km from north to south. The average annual precipitation is around 1400 mm. Rainfall is concentrated in the 5 month period from May to September.

Aquaculture is the primary source of revenue for the inhabitants in the coastal and plain region. A large amount of groundwater has been extracted from aquifers in this area to supply fishponds. Figure 1b displays the distribution of townships and classifies the aquaculturaral land-use into three categories, including primary aquacultural township (PAT), subordinate aquacultural township (SAT), and non-aquacultural township (NAT). They are defined by the ratio of aquacultural area to total township area. In PATs, more than 10% of the land is used for aquaculture; in SATs, between 2 and 10% of the land is used for aquaculture, and in NATs, less than 2% of the land is used for aquaculture.

Changes in sea level significantly influence the composition and structure of the geological environment in the shallow sedimentary basin of southwestern Taiwan (*20*). The sedimentary basin is formed by the alternating invasion and retreat of seawater with interlayered formations of marine and nonmarine sequences. Between 1992 and 2001, the Taiwan government implemented a long-term comprehensive groundwater monitoring network plan to explore the hydrogeological characteristics of the main groundwater system in this region. The hydrogeological study demonstrated that the area was formed in the late Quaternary age. Subsurface hydrogeological analysis to a depth of approximately 300 m shows that the formation can be clearly divided into six interlayered sequences in the coastal area of the Choushui River alluvial fan, including three marine sequences and three nonmarine sequences (*20*). The nonmarine sequences with high permeability can be considered to be aquifers. The marine sequences with fine sediments can be regarded as aquitards in which a relatively high As content is accumulated and deposited. However, the formation in the Chianan plain cannot be clearly partitioned into marine and nonmarine sequences because of the presence of many small aquitards.

The new groundwater monitoring network in the Choushui River alluvial fan and Chianan plain was quickly implemented. It consists of 56 hydrogeological investigation stations and 168 groundwater monitoring wells in aquifers of different depths (Figure 1a) (*21*). Most investigation stations have two to five monitoring wells, which are located in aquifers. The number of wells at a station depends on the number of aquifers.

Arsenic Concentrations in Groundwater. From 2000 to 2001, the Taiwan Sugar Company (*22*, *23*) undertook groundwater quality surveys in the Choushui River alluvial fan and in the Chianan plain. The surveys analyzed 31 water quality survey items, including the concentrations of As. An abnormally high As content was the only carcinogenic trace

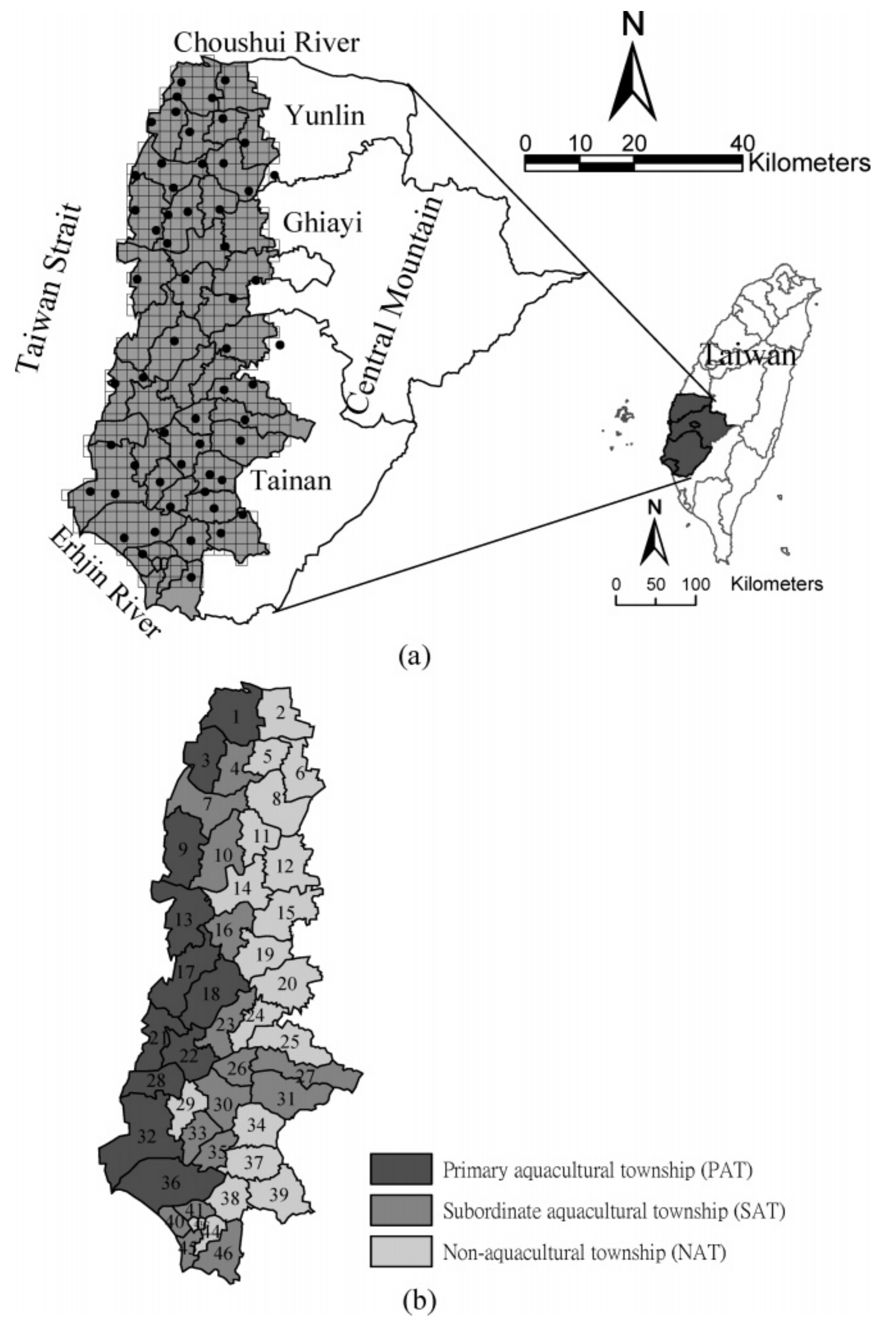

FIGURE 1. (a) Study site in southwestern Taiwan. The closed circles mark the locations of the hydrogeological stations. (b) Locations and number of various aquacultural townships.

element among these water quality items. Table 1 presents statistics concerning measured As concentrations in the study area. The average As concentration was 203.3 μg/L, with a maximum value of 1470 μg/L. Approximately 16% of the As concentrations in the surveys were below the detection limit of 10 μg/L, which is the regulation standard for drinking water in Taiwan. Figure 2 plotted the vertical distribution of the measured As concentrations. The high As-contaminated concentrations (>1000 μg/L) in groundwater were frequently found at elevations of −80 to −200 m.

Arsenic Exposure and Bioaccumulation Pathways. Water sources for aquacultural ponds are generally surface water and subsurface water adjacent to the ponds. The sources of surface waters, which have very low As concentrations, include river, irrigation and drainage channels, rain, and others. Arsenic of groundwater is the most primary As source in aquacultural ponds. The mass balance of As among groundwater, surface water, and pond water is expressed as follows:

$$C_{\mathrm{pond}} = \frac{Q_{\mathrm{well}}}{Q_{\mathrm{well}} + Q_{\mathrm{surface}}} C_{\mathrm{well}} = DR \times C_{\mathrm{well}} \qquad (4)$$

where Q_{well}, Q_{surface}, and Q_{pond} are the volumes of groundwater, surface water, and pond water, respectively. C_{well} and C_{pond} are the As concentrations in groundwater and pond water, respectively. The As concentration in surface water is considered to be none. The term $Q_{\mathrm{well}}/(Q_{\mathrm{well}} + Q_{\mathrm{surface}})$ is the dilution ratio (DR) regarding the use of surface water.

The As concentrations in fish is a result of bioaccumulation in fishpond environments and fish food. Fish bioaccumulation in ponds is called bioconcentration and that in fish food is called biomagnification (*24*). The artificial diets of

TABLE 1. Statistics Regarding Measured Concentrations of As in Groundwater[a]

statistics	As concentration
average	203.3
median	80
SD	291.7
minimum	<10
maximum	1470
Percentiles	
10th	<10
16th	10
20th	20
30th	30
40th	50
50th	80
60th	120
70th	153
80th	370
90th	621

[a] In μg/L.

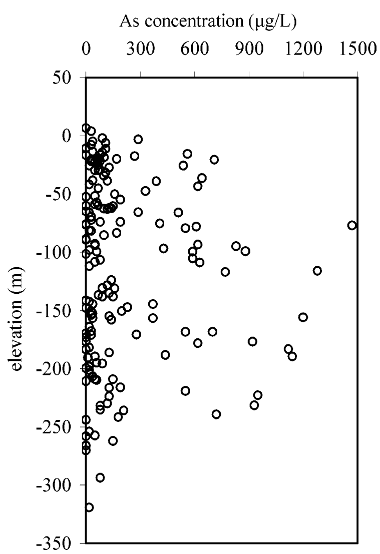

FIGURE 2. Vertical distribution of measured As concentrations. The zero elevation is the mean sea level.

cultivated fish do not contain As, resulting in that biomagnification can be ignored. Therefore, the As concentration in tilapia is expressed as follows:

$$C_{\text{tilapia}} = BCF \times C_{\text{pond}} \quad (5)$$

where C_{tilapia} is the As concentrations in tilapia. The BCF is the bioconcentration factor.

Eight sets of well water, pond water, and tilapia samples in four aquacultural ponds in the townships 17, 18, and 22 were surveyed from September to December 2002 (*25*). The well water, pond water, and tilapia samples were examined by atomic absorption spectrometry (AAS) to obtain their total As concentrations. Figure 3a plots the relationship between As concentrations in well water and in pond water. Additionally, Huang et al. (*26*) reported a detailed experimental analysis of tilapia, including 68 data on As concentrations in tilapia and 21 data on As concentrations in pond water in the townships 17, 18, 21, and 22. Three to four tilapia samples and one water sample were collected in a pond. This study combined all data regarding As concentrations of tilapia and pond water. Figure 3b plots the relationship between As concentrations in pond water and As concentrations in tilapia. Section B in our Supporting Information presents distributions of measured DR and BCF.

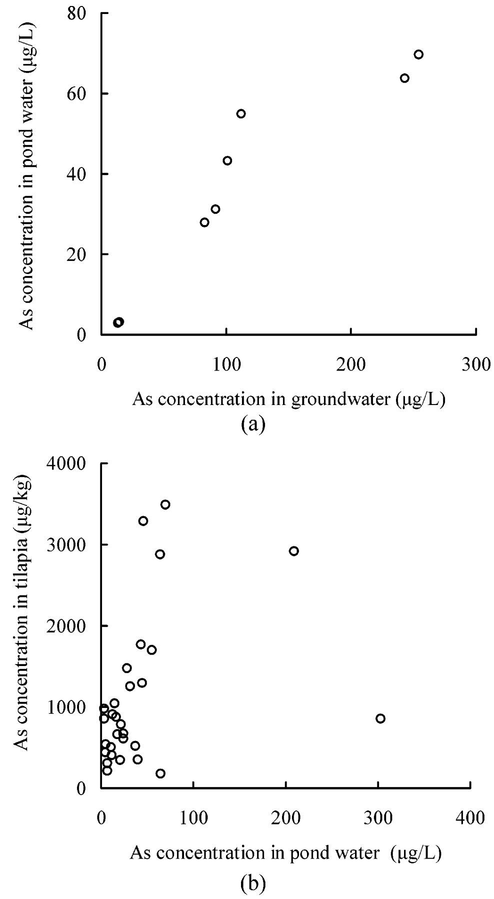

FIGURE 3. As concentrations (a) in well water and in pond water and (b) in pond water and in tilapia.

Human Health Risk Assessment. The Risk Assessment Forum reassessed the risk of cancer associated with the ingestion of inorganic As. The U.S. EPA Region III Risk-Based Concentration Table (*27*, *28*) supports a method for estimating the target cancer risk (TR). The risk of the carcinogenic effects of inorganic As is expressed as exceeding the probability of contracting the cancer over a lifetime of 70 years. A model for estimating the target cancer risks (lifetime cancer risks) is

$$\text{TR} = \frac{\text{EFr} \times \text{EDtot} \times \text{IR}_\text{t} \times C_t \times \text{CPSo}}{\text{BWa} \times \text{ATc}} \times 10^{-3} \quad (6)$$

where TR is the target cancer risk (the incremental individual lifetime cancer risk); EFr is the exposure frequency (350 days/year); EDtot is the exposure duration (30 years); IR_t is the ingestion rate in the edible portion of tilapia (g/day wet wt); C_t is the inorganic As concentration in the edible portion of tilapia (μg/g wet wt); CPSo is the oral carcinogenic potency slope (risk per (mg/kg/day)) (1.5 $(\text{mg/kg/day})^{-1}$); BWa is the body weight of a Taiwanese adult (60.9 kg) (*29*); and *ATc* is the averaging time for carcinogens (25 550 days). An estimated TR value of exceeding one millionth is typically considered to pose a potential threat to human health.

Eq 6 can be rewritten using the aforementioned parameters, the results of the experiment on tilapia, and the As exposure and bioaccumulation pathways.

$$\begin{aligned}\text{TR} &= \text{IR}_\text{t} \times C_t \times 10.13 \times 10^{-6} \\ &= (\text{IR}_\text{ft} \times \alpha) \times \left\{ \left[\frac{C_\text{well} \times \text{DR} \times \text{BCF}}{1000} \right] \times \beta \times (1 - w) \right\} \times 10.13 \times 10^{-6} \quad (7)\end{aligned}$$

TABLE 2. Used Parameters and Their Distributions on Risks of Cancer

parameters	values or distributions	estimation method	data sources
BWa (kg)	N(60.9, 9.9)[a]	constant (60.9)	Department of Health, Taiwan (*29*)
IR_{ft} (g/day wet wt)	5.4	constant (5.4)	Council of Agriculture, Taiwan (*30*)
α	N(0.32, 0.046)	variability (MC simulation)	Lin (*25*)
β	LN(0.05,2.71)[b]	variability (MC simulation)	Huang et al. (*26*)
w	N(0.773, 0.016)	variability (MC simulation)	Huang et al. (*26*)
DR	LN(0.329, 1.34)	variability (MC simulation)	Lin (*25*)
BCF	LN(29.1, 3.07)	variability (MC simulation)	Lin (*25*) and Huang et al. (*26*)
C_{well} ($\mu g/L$)	<10 to ∼1470[c]	spatial variability (SIS)	Taiwan Sugar Company (*22, 23*)

[a] N(μ_a,σ_a) denotes a normal distribution with an arithmetic average of μ_a and an arithmetic standard deviation of σ_a. [b] LN(μ_g,σ_g) denotes a log-normal distribution with a geometric average of μ_g and a geometric standard deviation of σ_g. [c] Minimum to ∼maximum.

where IR_{ft} is the ingestion rate of full tilapia (g/day wet wt); α is the edible ratio of tilapia; β is the ratio of inorganic As content to total As content in tilapia; and w is the water content of tilapia. Table 2 presents the variations and the distributions of these parameters used in eqs 6 and 7. Our experimental data (*25*) and the analytical results of Huang et al. (*26*) indicated that the arithmetic average α was 0.32 with the arithmetic standard deviation of 0.046; the arithmetic average w was 0.773 with the arithmetic standard deviation of 0.016, and the geometric average β was 0.05 with the geometric standard deviation of 2.71. According to the Taiwanese Food Supply and Demand Annual Report during 2002 (*30*), 85 143 tons of tilapia were produced, 43 670 tons were exported, and 40 tons were imported. The Taiwanese population over the age of 4 years was 21.07 million in 2002 (*31*). Consequently, the weight of tilapia ingested per Taiwanese per day, IR_{ft}, was 5.4 g/day wet wt. Section B in our Supporting Information exhibits distributions of observed data on the parameters. Parameters in eq 7 include three categories: (1) As exposure and bioaccumulation pathways (DR and BCF), (2) experimental data of tilapia (α, β, and w), and (3) As concentrations in groundwater (C_w). Because of sparse observed data, robust data concerning the parameters in the first two categories were generated using MC simulation based on their measured distributions and accounted for their uncertainty (*32*). The Risk (Version 4.5, Professional Edition, Palisade Corp.) software was used to analyze statistically the measured data and to carry out MC simulation.

Results and Discussion

Variogram Analysis and Realizations of As Concentrations. The measured As concentrations at the 16th, 30th, 40th, 50th, 60th, 70th, 80th, and 90th percentiles, as shown in Table 1, were used as the eight thresholds (z_k, $k = 1,2,..,8$). Section A in the Supporting Information presents the result of the analysis of the variograms.

Arsenic concentrations in groundwater shown in Figure 2 present three groups, <10−700 μg/L within elevations of 10 to −80 m (shallow layer), <10−1470 μg/L within elevations of −80 to −200 m (median layer), and <10−950 μg/L below the −200 m elevation (deep layer). Furthermore, a spatial pattern of high As-contaminated groundwater at the shallow layer is significantly different to those at the median and deep layers. Three layers at depths of 50, 150, and 250 m under the ground surface represented the use of groundwater for aqualculture at the shallow, median, and deep layers, respectively. Generally, groundwater at a depth of more than 200 m is less used and expensive for aquaculture, but it can provide a comparison with the other depths. One thousand realizations of As exposure concentrations in groundwater were reproduced using SIS to denote all possible outcomes. Each horizontal layer was divided into a grid of 21 × 49 cells, with a spacing of 2 km (Figure 1a). Section C in the Supporting Information shows one realization of As concentrations in groundwater and distributions of 1000 simulated data at some cells.

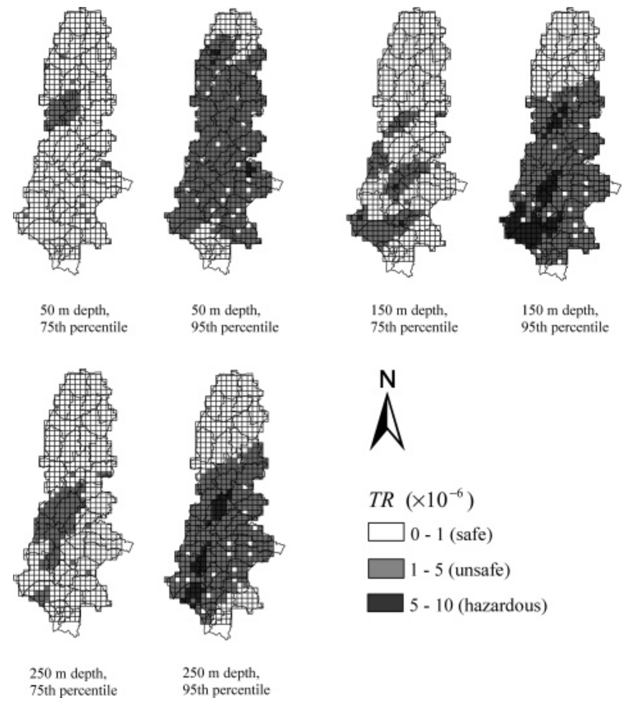

FIGURE 4. GIS maps of the TRs at the three depths.

Analysis of Uncertainty in Risk Assessment. A series of joint distributions integrating the aforementioned parameters were carried out. The As concentrations in groundwater, which is a nonparametric distribution, for each cell were combined in the last process of the joint distributions. Traditionally, the 5th, 25th, 50th, 75th, and 95th percentiles of risk are displayed in a box-and-whiskers plot to assess different likelihoods of exceeding risk levels (*7*, *8*). In this work, the five percentiles of TR at each cell were determined to assess spatially health risks under different probabilities. Section B in the Supporting Information reveals the detailed processes of the joint distributions and risk assessment.

Carcinogenic Risks Associated with Ingesting Arsenic in Aquacultural Tilapia. Irregular variations present at the TR maps are due to the highly heterogeneous characteristics of 1000 As realizations reproduced by SIS. All TRs determined from the 5th and 25th percentiles are below one millionth; few TRs determined from the 50th percentile exceed one millionth, but massive TRs determined from the 75th and 95th percentiles exceed one millionth at the cells of the three depths. Figure 4 shows cell-level GIS (geographic information system) maps of the 75th and 95th percentiles of TR at the three different depths. Three levels of TR, $0{-}1 \times 10^{-6}$ (safe), $1{-}5 \times 10^{-6}$ (unsafe), and $5{-}10 \times 10^{-6}$ (hazardous), are displayed. A higher level of TR corresponds to a higher menace to human health and means a more strict control on groundwater use. At the 50th percentile of risk, only 1.5% of TRs exceed one millionth, and the largest TR is 2.49×10^{-6}. At the 75th percentile of risk, 16.3% of TRs exceed one millionth, and the largest TR is 6.24×10^{-6}. At the 95th percentile of risk, 70.9% of TRs exceed one millionth, and the largest TR is 7.76×10^{-6}. The largest TRs all occur at the 150 m depth.

The 95th percentile of risk is by construction always greater or equal to the 75th percentile of risk, and the unsafe and hazardous regions at the 95th percentile of risk are extensive throughout most regions. Thus, this study suggests that the 75th percentile of health risk is adopted to assess the risks of cancer associated with ingesting As contents in aquacultural tilapia in performing the preliminary remedial framework. The 95th percentile of health risk computed from extrapolating ccdfs using a hyperbolic model and quantifying parameter uncertainty using a MC simulation is considered with extreme caution of maximally exposed risks. In terms of the spatial distributions of health risks, the risk pattern at the 50 m depth overtly differs from those at the 150 and 250 m depths. At the 50 m depth, the regions with high risks are concentrated in the central coastal areas. At the 150 and 250 m depths, the regions with high risks are situated in the central and southern coastal areas and the central inner areas. Furthermore, the potential risks of cancer are emphasized in PATs and SATs. For the 75th percentile of TR, the health risks of cancer are unsafe or hazardous for the PAT 13 and the SAT 16 at the 50 m depth, for the PATs 17, 21, 22, 32, and 36 and the SATs 16, 23, 26, 30, 33, and 35 at the 150 m depth, and for the PATs 17, 18, 21, 22, and 36 and the SAT 23 at the 250 m depth. The groundwater used in the northern townships at the 150 and 250 m depths are safe for farming tilapia at the 75th and 95th percentiles of risk.

Han et al. (*7*) reported a maximal risk of 65.6×10^{-6} and a typical risk of 26.2×10^{-6} for ingesting As contents in farmed tilapia in the BFD areas of Taiwan. Ling et al. (*33*) documented that the 90th percentile of TR associated with ingesting As contents in farmed tilapia in the BFD areas ranged from 0.015×10^{-6} to 1.62×10^{-6} for city residents and from 2.07×10^{-6} to 78.9×10^{-6} for subsistence fishers. The health risks estimated in our study fall into the range of Ling et al. (*33*) but are smaller than the result of Han et al. (*7*). The discrepancy of the evaluated results is that many evaluated parameters and methods used in risk assessment are different among our and their researches.

In conclusion, this work spatially analyzed the potential risks of cancer associated with ingesting As in aquacultural tilapia in the coastal regions of southwestern Taiwan, where BFD prevails. Owing to sparse measured data, MC simulation and SIS properly accounted for uncertainty of parameters and can be used to establish a sound framework of spatial risk analysis. The probabilistic risk assessment provided more useful information than the deterministic one, which poorly propagated the uncertainty of parameters. The probabilistic outcomes formulated proper strategies under various remedial phases. The 75th percentile of risk should be considered in the preliminary phase of remediation. This investigation focuses on the use of groundwater with various levels of As exposure, which induces the potential risk of cancer associated with consuming aquacultural tilapia. Therefore, if the TRs in the PATs and SATs exceed one millionth, the use of groundwater in aquaculture should be reduced or prohibited, or the depths at which groundwater are withdrawn should be changed. Furthermore, regarding the development of aquacultural businesses in NATs, this work provides a reference for determining whether groundwater can be used as a main water source to meet the needs of aquaculture. When enough surface water is available to meet the aquacultural needs of BFD regions, health risks should be evaluated more strictly using the 95th percentile of risk.

Acknowledgments

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Note Added after ASAP Publication

The genus species name for Tilapia was misspelled in the version published ASAP January 24, 2006; the corrected version was published ASAP on February 7, 2006.

Supporting Information Available

Detailed analysis and the results of 3-D anisotropic variability, analysis of uncertainty in risk assessment, and As realizations in groundwater. This material is available free of charge via the Internet at http://pubs.acs.org. Spatially potential carcinogenic risks for arsenic contents in farmed tilapia via a bioaccumulation pathway in blackfoot disease hyperendemic regions of southwestern Taiwan are presented.

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