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RESEARCH ARTICLE

Coupling phytotoxicity and human health risk assessment to refine the soil quality standard for As in farmlands

Kai-Wei Juang[1] · Li-Jia Chu[1,2] · Chien-Hui Syu[3] · Bo-Ching Chen[2]

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Abstract

In the present study, a field experiment was conducted to investigate arsenic (As) concentrations in soils and in grains of 15 rice varieties in a contaminated site in Taiwan. The studied site was divided into two experimental units, namely plot A and plot B. The results showed that mean total As concentrations were 70.94 and 61.80 mg kg^{-1} in plot A and plot B, respectively, and thus greater than or approximate to the soil quality standard for total As in Taiwan (60 mg kg^{-1}). The As levels in rhizosphere soil in plot A (19.71–32.33 mg kg^{-1}) were much higher than in plot B (6.41–8.60 mg kg^{-1}); however, As accumulation in brown rice did not significantly differ between the plots. These results implied that a significant variation in the bioconcentration factor (BCF) value of As existed among different rice genotypes, and a negative correlation was observed between BCF value and rhizosphere As level in the soil. In phytotoxicity, the median values of the ecological risk indicator were 104.85 and 103.89 in plot A and plot B, respectively, indicating considerable risk. In human health risk assessment, the median and 97.5%-tile values for cancer risk for both male and female residents were markedly higher than the acceptable risk (1×10^{-4}). Furthermore, non-cancer and cancer risks were higher for males than females, mainly due to the greater rice ingestion rate of males. Sensitivity analysis showed that total As concentration in soil was the main factor affecting health risks, suggesting that priority should be given to the reduction of soil As levels. To better manage the phytotoxicity of As on rice, as well as the health risk to residents resulting from exposure to As-contaminated soils, the soil quality standard for As in farmland soils should be set between 5 and 10 mg kg^{-1}. The methodology developed in this study could also be applied to provide the basis for refining and revising the soil quality standard for heavy metals in farmland in other regions and countries.

Keywords Arsenic · Bioconcentration factor · Rice · Risk · Soil quality standard

Introduction

As is ubiquitous in the ecosystem through natural routes, such as weathering of minerals and denudation of bedrock, or through anthropogenic activities such as mining, metal processing and smelting, chemical production, burned vegetation, and transport emission (Duan et al. 2017; Alexakis 2020; Sarwar et al. 2021; Zhang et al. 2021; Painecur et al. 2022). At a world level, it is reported that the background level of total As in crust and soil is 1.8 and 5–6 mg kg^{-1}, respectively; however, the As level in the soil is highly correlated with the land use (Liu et al. 2004; Alexakis and Gamvroula 2014). For instance, Alexakis et al. (2021) investigated the spatial distribution of As content in the soil of the Ioannina basin and reported that the maximum As level in soil is 76, 27, and 33 mg kg^{-1} for agricultural land use, urban land use, and wetlands, respectively. Additionally, long-term irrigation of As-contaminated groundwater has increased As levels in many agricultural soil environments (Majumder and Banik 2019). For example, the groundwater As level could reach 2 mg L^{-1} in some areas of Bangladesh, where the WHO-permissible limit for drinking water is only

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Bo-Ching Chen
bcchen@nhu.edu.tw

[1] Department of Agronomy, National Chiayi University, Chiayi, Taiwan

[2] Department of Natural Biotechnology, Nanhua University, 622 No. 55, Sec. 1, Nanhua Rd., Dalin Township, Chiayi, Taiwan

[3] Agricultural Chemistry Division, Taiwan Agricultural Research Institute, Taichung, Taiwan

0.01 mg L^{-1}. As a consequence, the As levels in soils could reach up to 83 mg kg^{-1} in areas irrigated with contaminated water in Bangladesh (Abedin et al. 2002).

Owing to its persistence, high bioaccumulation, and toxicity in farmland ecosystems, As pollution has become a serious problem worldwide. When excess As enters agricultural soil, it may cause a decrease in soil productivity and threaten human health. Arsenic exposure can induce various cardiovascular, neurological, respiratory, and other systemic diseases (Juang et al. 2021; Sarwar et al. 2021). Among different As species, inorganic arsenic (iAs) has been classified as a class A human carcinogen that can cause lung, skin, liver, bladder, and kidney cancers (Duan et al. 2017; Sarwar et al. 2021). Consequently, the phytotoxicity as well as the human health risk of As in farmland ecosystems has become a major global concern and has been extensively studied in the past few decades (Juhasz et al. 2006; Lorenzana et al. 2009; Rasheed et al. 2016; Bradham et al. 2018; Barcelos et al. 2020).

By definition, ecological risk assessment is the process of evaluating how likely it is that the environment might be impacted as a result of exposure to one or more environmental stressors (US EPA 1992). In recent years, practical indexes such as the geoaccumulation index (I_{geo}), single factor pollution index (NCPI), and ecological risk index (E_r) have been recommended for quantitative evaluation of the toxic risk of heavy metal in an environmental medium (Huang et al. 2019; Xiao et al. 2019; Prabagar et al. 2021; Lü et al. 2022). Among these indexes, the ecological risk index has been adopted by numerous researchers because it is easier to compute and takes both actual and background levels of metal into consideration. For instance, Prabagar et al. (2021) studied the levels of seven heavy metals in grapevine soils and applied the ecological risk index to assess the impact of these metals on the studied soils. However, few studies have attempted to incorporate the ecological risk index into phytotoxicity and health risk assessment to comprehensively understand the influence of the studied metals on both the ecosystem and human health (Xiao et al. 2019; Zhang et al. 2021).

Rice (*Oryza sativa* L.) is a staple food for more than 3 billion people worldwide. The consumption of rice and rice products has been recognized as the primary source of As exposure, especially the more deleterious iAs form (Chi et al. 2018; Samal et al. 2021). The migration and accumulation of As in rice might be influenced by two dominant factors, namely rice genotype and the physicochemical properties of soil (Juang et al. 2021). Numerous studies have proposed that significant differences in As accumulation in grains exist for different rice genotypes, even those cultivated in the same rice paddy field (Carey et al. 2010; Chen et al. 2016; Juang et al. 2021). Ma et al. (2008) further indicated that the As level in grain may exceed the regulatory standard for some genotypes of rice cultivated in paddy soils containing background or low As concentration. On the other hand, physicochemical properties including soil texture, pH, organic matter, and cation exchange capacity may affect the fractionation of As in soil, thus influencing the bioavailability and accumulation of As in rice grain (Pérez-Sirvent et al. 2007). When assessing the transport AQ3 and accumulation of As in the soil–rice system, therefore, it is necessary to take bioavailability into consideration.

Traditionally, total As concentrations in grains have been used to estimate the daily intake associated with rice consumption; however, this may result in the overestimation of exposure dose due to a lack of consideration of As bioaccessibility (Li et al. 2017; Liu et al. 2017; Sharafi et al. 2019). Bioaccessibility is defined as the fraction that can be transformed into absorbable forms during digestion (Liu et al. 2017; Guo et al. 2022). Compared with total concentration, bioaccessible As concentration is more suitable and realistic for assessing actual human health risks associated with rice consumption (Sharafi et al. 2019; Wang et al. 2021; Guo et al. 2022). Recently, many researchers have studied the bioaccessibility of As to better understand As accumulation and toxicity in the rice–human system. For example, Omar et al. (2015) investigated the bioaccessibility of nine heavy metals and indicated that nonessential elements such as As may have lower bioaccessibility than essential microelements. Li et al. (2017) studied 55 rice samples collected from a large geographic distribution across China and suggested that a suitable default value of 39.9% might be adopted for bioaccessible rice iAs. Li et al. (2021) further suggested that As bioaccessibility is highly variable among rice genotypes. For As species, As (III) is proposed to be the dominant bioaccessible species, followed by As (V), DMA, and MMA (Wang et al. 2021). To accurately determine the health risk of rice consumption, therefore, the inclusion of bioaccessibility in the human health risk assessment paradigm is critical.

Despite the fact that As is harmful to ecosystems and human beings, most research and regulatory standards regarding the risk of As in paddy soils have focused solely on phytotoxicity or human health risk. Moreover, most previous studies have employed total As concentration to estimate the potential health risk posed by As in the rice grain. As mentioned, lack of consideration of As bioavailability and bioaccessibility may reduce the accuracy of human health risk assessment. In light of these considerations, a field experiment was conducted in the present study to investigate As levels in the soil and the corresponding rice grain in a rice paddy field in Taiwan. The objectives of the study were (1) to understand the influence of biogeochemical character on the bioconcentration of As in the paddy soil, (2) to evaluate the phytotoxicity of As on rice based on the ecological risk index, (3) to assess the comprehensive health

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risk posed by As in the studied soil by taking both bioconcentration and bioaccessibility into consideration, and (4) to examine the current soil quality standard for As through a retrospective health risk assessment.

Materials and methods

Field experiment and sampling

A field experiment was conducted on a rice paddy farm located in central Taiwan from August to December 2020 (Fig. 1). Based on the results of our preliminary investigation, the soil texture on the studied farm was classified as sandy loam. The pH, electrical conductivity, and organic matter (OM) of the soil were 7.17, 0.41 dS m^{-1}, and 3.05%, respectively (Juang et al. 2021). According to their distance from the irrigation well, the site was divided into two experimental units: one was the “plot A” unit, which was closer to the irrigation well, and the other was the “plot B” unit, which was located a distance from the irrigation well. The cultivars were arranged in these two plots in a strip-plot design proposed by Milliken et al. (1998). Each plot was further divided into 15 blocks; each block contained three planting lines for the cultivation of rice. Fifteen rice cultivars popular among local residents were selected for the experiment: TK2, TK9, TK14, TK16, TY3, TNG71, TC192, TN11, KH139, TT30, TCS10, TKW3, TCSW2, TCS17, and KHS7. During the experimental period, the irrigation frequency was twice a week and was adjusted according to rainfall. For fertilizer management, the soils were supplemented with 150 kg N ha^{-1} as $(NH_4)_2SO_4$, 40 kg P_2O_5 ha^{-1} as KH_2PO_4, and 90 kg K_2O ha^{-1} as K_2SO_4. At the end of the field experiment (December 2020), three plants for each cultivar were randomly selected and harvested from each plot, and the corresponding rhizosphere soils were also collected. Two sets of parallel experiments were conducted; thus, each cultivar had six replicates.

Sample processing and chemical analysis

All the collected samples were transported to a laboratory in sealed polyethylene bags. Grain samples were thoroughly washed with distilled water, oven-dried at 75 °C, and then unhusked to obtain brown rice samples. Meanwhile, the soil samples were air-dried at room temperature and then ground to pass through a 2-mm sieve. The brown rice and soil samples were then kept in polyethylene bags until analysis.

To determine the total, rhizosphere, and exchangeable As, soil samples were extracted with HNO_3–H_2O_2 mixture, DCB (dithionite-citrate-bicarbonate) solution, and NaH_2PO_4, respectively. The rhizosphere As was generally regarded as the fraction of total As that can be potentially absorbed by the rice root surface (Wang et al. 2018). On the other hand, the exchangeable As was regarded as the fraction of total As that can be fast absorbed by plants (Alarm and Tokunaga 2006). Brown rice samples were ground and then digested completely with 0.28 M HNO_3 at 95 °C for 90 min (Yao et al. 2021). The As concentration in digests and extracts for soil samples was then determined with an inductively coupled plasma-mass spectrometer (ICP-OES, Agilent 7700×) (Juang et al. 2021). The extraction method for determining As species in plant samples was modified by Ma et al. (2017). The As species, including As (III), As

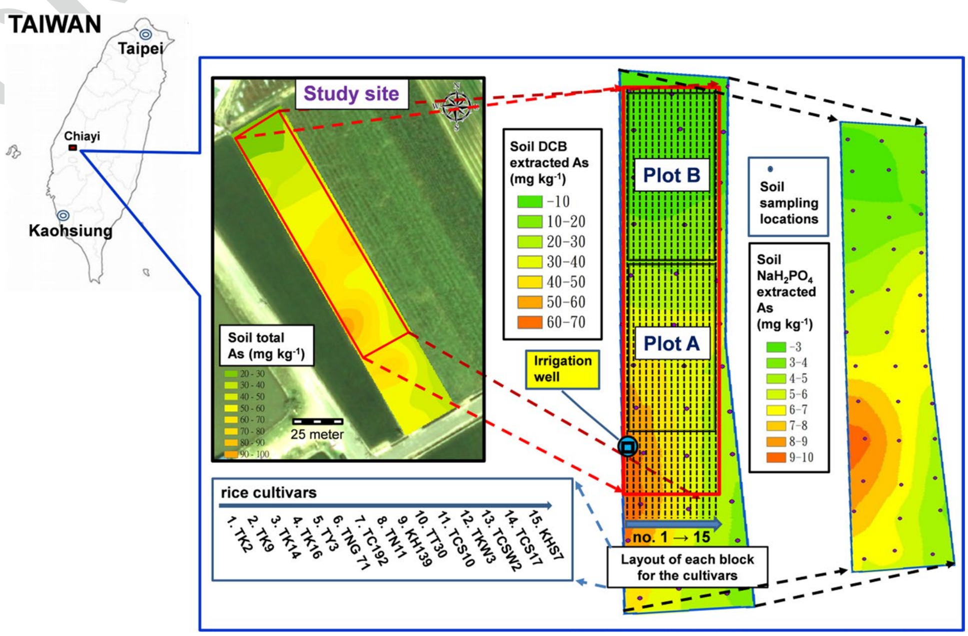

Fig. 1 Location map, soil sampling configuration, and spatial distributions of the total, DCB-extracted, and NaH_2PO_4-extracted As concentration in soils of the studied site

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(V), DMA, and MMA, in each extract, were determined by using HPLC-ICP-MS with the equipment parameters proposed by Liao et al. (2021). Meanwhile, Certified Reference Material (CRM) ERM BC-211 was analyzed together with rice samples.

Rhizosphere As fraction

Rhizosphere As fraction (f_{rhizo}, %) was calculated as follows:

$$f_{\mathrm{rhizo}} = \frac{C_{\mathrm{soil,rhizo}}}{C_{\mathrm{soil}}} \tag{1}$$

where C_{soil} (mg kg^{-1}) is the total As concentration in the soil; $C_{\mathrm{soil,rhizo}}$ (mg kg^{-1}) is the As concentration in the rhizosphere soil.

Bioconcentration factor

Bioconcentration factor (BCF) is defined as the ability of paddy rice to absorb and retain As from rhizosphere soil and can be calculated as follows:

$$BCF = \frac{C_{\mathrm{rice}}}{C_{\mathrm{soil,rhizo}}} \tag{2}$$

where C_{rice} (mg kg^{-1}) is the As concentration in brown rice.

Phytotoxicity of As on rice

To evaluate the phytotoxicity of As on rice, the ecological risk index (E_r) proposed by Hakanson (1980) was applied. It can be calculated as

$$E_r = \frac{C_{\mathrm{soil}}}{C_{\mathrm{b}}} \times T \tag{3}$$

where C_{soil} is the measured total As concentration in soil samples (mg kg^{-1}), C_{b} represents the background value of total As in soil, and T is the toxic factor of As. Chang et al. (1999) investigated As content in surface soils (0–15 cm) in Taiwan and proposed that the mean As concentration in agricultural soils is 5.65 mg kg^{-1}. In addition, the T value for As is 10, according to Hakanson (1980). The ecological risk index, E_{r}, is classified into five categories: low risk ($E_{\mathrm{r}} < 40$), moderate risk ($40 \le E_{\mathrm{r}} < 80$), considerable risk ($80 \le E_{\mathrm{r}} < 160$), high risk ($160 \le E_{\mathrm{r}} \le 320$), and very high risk ($E_{\mathrm{r}} > 320$) (Huang et al. 2019; Xiao et al. 2019; Prabagar et al. 2021; Zhang et al. 2021).

Human health risk assessment

The human health risks of iAs associated with rice consumption by Taiwan residents were determined following the guidance proposed by the US EPA (US EPA 1992; 2002) with modifications. In the exposure assessment, the estimated daily intake (EDI, mg kg^{-1} d^{-1}) of iAs in rice was calculated as follows:

$$\mathrm{EDI} = \frac{C_{\mathrm{soil}} \times f_{\mathrm{rhizo}} \times BCF \times IR \times P_{\mathrm{inorg}} \times P_{\mathrm{white/brown}} \times BAc \times CF}{\mathrm{BW}} \tag{4}$$

where IR is the ingestion rate of brown rice (g d^{-1}). P_{inorg} represents the iAs proportion of total As in brown rice; $P_{\mathrm{white/brown}}$ represents the proportion of iAs in white rice relative to that in brown rice. BAc is the ratio of iAs that can be absorbed into the systemic circulation following consumption of iAs in white rice (i.e., bioaccessibility). CF is the conversion factor (kg g^{-1}), and BW is the body weight of the considered population (kg).

Due to the high variability and uncertainty of the studied population, some parameters in Eq. (4) were treated probabilistically. Considering the restrictions on the domain of the variables, some parameters, including C_{soil}, BCF, and IR, were transformed from normal distribution to lognormal distribution to avoid errors (i.e., negative values) in the simulation. Since IR and BW in Eq. (4) are quite different according to sex, the health risk of female and male populations was estimated separately. As a consequence, the distribution of EDI was obtained from the simulation result of Eq. (4). Then, the median and 97.5%-tile value of EDI was used to express central tendency exposure (CTE) and the plausible worst-case reasonable maximum exposure (RME), respectively.

In risk characterization, both carcinogenic and noncarcinogenic risks were considered. For noncarcinogenic risk, the hazard quotient (HQ), representing the ratio of EDI to the reference dose (RfD, mg kg^{-1} d^{-1}) of iAs, was calculated as follows:

$$\mathrm{HQ} = \frac{\mathrm{EDI}}{\mathrm{RfD}} \tag{5}$$

The inherent assumption of HQ is that there is a threshold of exposure below which it is unlikely that the considered population will experience adverse health effects. If the HQ exceeds unity, potential noncarcinogenic effects might be a concern (Juang et al. 2021).

For carcinogenic risk, the target cancer risk (TR), representing the probability of an individual developing cancer over a lifetime as a result of exposure to a potential carcinogen, was calculated as follows:

$$\mathrm{TR} = \mathrm{EDI} \times \mathrm{SF} \tag{6}$$

where SF is the slope factor of iAs (kg d mg^{-1}). If $\mathrm{TR} < 10^{-6}$, the carcinogenic risk is considered negligible; if $\mathrm{TR} > 10^{-4}$, the risk is considered unacceptable by most international regulatory agencies. According to the US EPA's guidance,

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the carcinogenic risk is considered acceptable or tolerable if $10^{-6} < TR < 10^{-4}$ (Sharafi et al. 2019).

Statistical and uncertainty analysis

All experimental data were analyzed using Microsoft Excel 2013 and represented as the mean or mean with standard deviations for several samples of soils or each genotype of rice. Statistica software (Ver. 13.3, TIBCO Software Inc.) was employed to generate box-whisker plots. In addition, Monte-Carlo simulation with 10,000 iterations was performed using Oracle Crystal Ball software (Ver. 11.1.2.4.850, Oracle®) to estimate E_r, EDI, HQ, and TR considering the distribution of independent variables (C_{soil}, f_{rhizo}, BCF, IR, BAc, and BW) in Eqs. (3) and (4).

Results and discussion

As accumulation in soils and rice grains

The total As concentrations in plot A and plot B were 70.94 ± 39.17 (mean ± standard deviation, $n = 19$) and 61.80 ± 18.72 mg kg^{-1}, respectively, which was greater than or near the soil quality standard for total As in farmland soil used for food crop production in Taiwan (i.e., 60 mg kg^{-1}) (EPA-TW 2011). A larger variation of As concentration was found in plot A than in plot B (Fig. 1). In addition, the As levels accumulated in rhizosphere soil in plot A (19.71–32.33 mg kg^{-1}) was much higher than in plot B (6.41–8.60 mg kg^{-1}) (Table 1), which was likely because the sampling sites in plot A were much closer to the irrigation well than those in plot B. Interestingly, As accumulation in rice grains did not significantly differ between the two plots. The total As concentrations in grains ranged from 0.19 to 0.47 mg kg^{-1} for plot A and from 0.22 to 0.45 mg kg^{-1} for plot B, which were higher than the local (0.117–0.216 mg kg^{-1}) and global (0.08–0.2 mg kg^{-1}) normal range reported by previous studies (Chen et al. 2016; Majumder and Banik 2019). On the other hand, since BCF was defined as the ratio of As level in rice grain to that in soil, BCF in plot A (0.0076–0.0172) was relatively lower than that in plot B (0.0253–0.0590) (Table 1 and Fig. 2).

By comparing the total concentration with the rhizosphere concentration of metals, it is generally recognized that the latter is a better indicator to relate metal accumulation in plants and available metal content in soils (Liu et al.

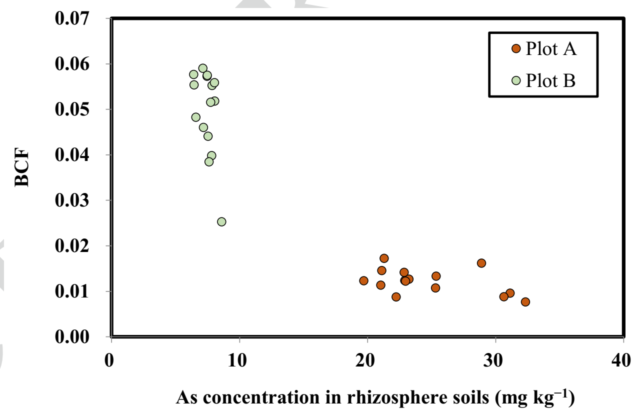

Fig. 2 Relationships between bioconcentration factor (BCF) and As concentration in rhizosphere soils at the studied sites

Table 1 As concentration in rhizosphere soils, total As concentration in grains, and bioconcentration factor (BCF) of different rice genotypes cultivated in plot A and plot B

Cultivar	Plot A			Plot B		
	Rhizosphere As (mg kg^{-1})	Total As in grain (mg kg^{-1})	BCF	Rhizosphere As (mg kg^{-1})	Total As in grain (mg kg^{-1})	BCF
TK2	31.13	0.30	0.0096	7.46	0.43	0.0572
TK9	32.33	0.25	0.0076	6.44	0.36	0.0554
TK14	30.64	0.27	0.0088	6.41	0.37	0.0577
TK16	22.85	0.32	0.0142	6.58	0.32	0.0483
TY3	22.90	0.28	0.0124	8.05	0.42	0.0518
TNG71	28.89	0.47	0.0162	7.13	0.42	0.0590
TC192	25.31	0.27	0.0107	7.17	0.33	0.0460
TN11	25.36	0.34	0.0133	7.54	0.33	0.0441
KH139	21.10	0.31	0.0146	7.84	0.43	0.0552
TT30	23.23	0.29	0.0127	8.60	0.22	0.0253
TCS10	21.03	0.24	0.0113	7.81	0.31	0.0398
TKW3	22.96	0.28	0.0122	8.04	0.45	0.0559
TCSW2	21.29	0.37	0.0172	7.47	0.43	0.0575
TCS17	19.71	0.24	0.0123	7.61	0.29	0.0384
KHS7	22.23	0.19	0.0087	7.74	0.40	0.0515

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2017; Xiao et al. 2019). In the present study, however, dissimilar results were observed between the As concentration in rice grain and the corresponding content in rhizosphere soils. The bioavailability of As is influenced by many soil properties, such as pH, cation exchange capacity (CEC), and OM (Gao et al. 2021). In this study, a relatively higher rhizosphere As fraction in plot A may be due to a long-term reduction in paddy soil condition caused by higher soil water content, thus favoring As (V) transformation to mobile As (III). On the other hand, an increase in soil water content enhances the formation of Fe-plaque in plant roots due to the dissolution of iron oxides in rhizosphere soils. The formation of Fe-plaque is generally regarded as a buffer that can restrict As entry into roots (Liu et al. 2004; Juang et al. 2021; Khanam et al. 2022). More As will be absorbed by Fe-plaque and reduce mobility to roots when the soil water content is high, overriding the effect of its increased availability. Therefore, the proportion of As that is actually absorbed by roots is lower in plot A than in plot B.

It is generally recognized that most of the As absorbed by rice plants remained in the root (Juang et al. 2021). Following the entry into the root, the translocation of As from root to grain is controlled by the abundance of nodes in shoots, As transporters and its chelating substances, and genes associated with As transport and binding (Gao et al. 2021; Khanam et al. 2022). Consequently, As transport to and accumulation of brown rice will vary among rice cultivars. In the present results (Table 1 and Fig. 2), all BCF values were less than 0.1, revealing that the uptake and translocation of As from soil to grains are limited. Additionally, BCF values varied dramatically among cultivars at relatively lower accumulated-As levels in the soil in plot B. With regard to genotypic difference, it is generally recognized that Japonica cultivars have lower As accumulation and translocation rates from straw to grain than Indica cultivars (Mridha et al. 2022). In the present results, however, the three highest BCF values were found for Japonica cultivars (TNG71, TK14, and TK2), whereas the lowest BCF values were found for Indica cultivars (TCS17 and TCS10), with the exception of TD30. Syu et al. (2014) indicated that the translocation factor from root to shoot of Japonica cultivars is significantly higher than that of Indica cultivars. The higher BCF value for Japonica cultivars observed in this study may thus be attributed to the higher translocation rate of As from root to shoot, with its ultimate accumulation in grains. On the other hand, BCF values were fairly low and remained nearly stable, with a relatively higher accumulated-As level in plot A. These results imply that, at low accumulated-As levels in the soil (i.e., plot B), a large proportion of As could be absorbed by roots, and then transported to grains because the biotic ligands (e.g., nodes in shoots and As chelating substances) within rice plants remained unsaturated. Consequently, the translocation of As from roots to grains was dominated by the rice genotype. Furthermore, in comparison to Indica cultivars, Japonica cultivars were more prone to accumulating As in grains by bioconcentration. At a high accumulated-As level (i.e., plot A), however, the absorption and translocation of As will be constrained because the biotic ligands within rice plants were nearly occupied and saturated. It can thus be speculated that different adaptation strategies were adopted by the rice when exposed to different ranges of available As in soil.

Among the different As species, the dominant one in brown rice was As (III), followed by DMA. The level of As (V) was relatively low, whereas MMA was nearly undetectable in the present study (Table 2). The levels of iAs in different rice cultivars varied, being 0.15–0.25 and 0.18–0.27 mg kg^{-1} in plot A and plot B, respectively. According to the maximum allowable level of iAs in brown rice set by the local government (i.e., 0.2 mg kg^{-1}), approximately 60% of rice cultivars (9 of 15) in plot A and 87% of rice cultivars (13 of 15) in plot B exceeded the regulatory standard. No significant difference was observed for the mean proportion of iAs in total As in brown rice (P_{inorg}) between plot A and plot B. The mean percentage of P_{inorg} was 68.36%, which was similar to that reported in previous research (Sun et al. 2008; Juang et al. 2021). On the other hand, an obvious genotypic difference for P_{inorg} was observed. The P_{inorg} value ranged from 0.53 to 0.92 and from 0.53 to 0.91 in plot A and plot B, respectively.

Levels of inorganic As in rice have received increasing attention, as iAs is highly toxic to human beings. Consequently, some national legislative limits for As in rice have been set based on iAs, rather than total As, for the protection of human health (Juang et al. 2021). In paddy soils, As (III) is the predominant As species because of their long-term flooded (anaerobic) conditions (Syu et al. 2014; Mridha et al. 2022). In rice grains, however, the proportions of As species are variable and strongly dependent on the rice genotype and on physicochemical and environmental interactions (Kumarathilaka et al. 2018). For instance, it was reported that inorganic As is predominant in Asian rice, whereas DMA is the most dominant species in rice produced in Europe and the USA (Islam et al. 2016; Mridha et al. 2022). A consistent result was also obtained in this study since all P_{inorg} values were greater than 0.5. The relatively higher value for P_{inorg} may also be attributed to the rice type analyzed in this study since brown rice generally contains a higher proportion of iAs as compared to polished rice (Majumder and Banik 2019). On the other hand, Wu et al. (2011) proposed that the proportion of DMA increases with increasing soil As content. Moreover, Zavala et al. (2008) indicated that DMA is the major species with relatively higher As levels in rice. In the present results, however, the influence of As content in soils and rice on the proportions of iAs and DMA in

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Table 2 Arsenite (As (III)), arsenate (As (V), DMA), total As concentration, and the proportion of iAs of total As (P_{inorg}) in brown rice of different rice genotypes cultivated in plot A and plot B

Cultivar	Plot A					Plot B				
	As (III)	DMA	As (V)	Total As	P_{inorg}	As (III)	DMA	As (V)	Total As	P_{inorg}
	(mg kg^{-1})					(mg kg^{-1})				
TK2	0.17	0.11	0.01	0.30	0.60	0.21	0.20	0.02	0.43	0.53
TK9	0.20	0.02	0.03	0.25	0.92	0.23	0.10	0.03	0.36	0.72
TK14	0.18	0.07	0.02	0.27	0.74	0.22	0.14	0.01	0.37	0.62
TK16	0.19	0.12	0.01	0.32	0.63	0.19	0.13	0.00	0.32	0.59
TY3	0.20	0.08	0.01	0.28	0.75	0.21	0.19	0.02	0.42	0.55
TNG71	0.23	0.22	0.02	0.47	0.53	0.24	0.15	0.03	0.42	0.64
TC192	0.19	0.07	0.01	0.27	0.74	0.22	0.09	0.02	0.33	0.73
TN11	0.20	0.14	0.00	0.34	0.59	0.20	0.14	0.00	0.33	0.61
KH139	0.18	0.12	0.01	0.31	0.61	0.22	0.20	0.02	0.43	0.56
TT30	0.17	0.11	0.02	0.29	0.66	0.19	0.02	0.01	0.22	0.91
TCS10	0.17	0.07	0.01	0.24	0.75	0.21	0.08	0.02	0.31	0.74
TKW3	0.19	0.08	0.01	0.28	0.71	0.22	0.21	0.02	0.45	0.53
TCSW2	0.22	0.15	0.00	0.37	0.59	0.24	0.16	0.03	0.43	0.63
TCS17	0.14	0.10	0.01	0.24	0.63	0.17	0.11	0.01	0.29	0.62
KHS7	0.17	0.02	0.00	0.19	0.89	0.21	0.19	0.00	0.40	0.53

brown rice was negligible. This contradictory result may be due to various factors, such as the difference in experimental conditions, soil environments, and rice genotypes. From the perspective of health risk, three rice genotypes, namely, TK16, TD30, and TCS17, are recommended in this study because they accumulated stable-low levels of iAs in grains in both plots.

Phytotoxicity assessment

To quantitatively express the ecological impact of As in the studied sites, *Er* values were calculated. The results are shown in Fig. 3. The median *Er* values in plot A and plot B were 104.85 and 103.89, respectively, indicating that in average situations, As in soil would pose a considerable risk to rice in both plots. More specifically, approximately 23.76

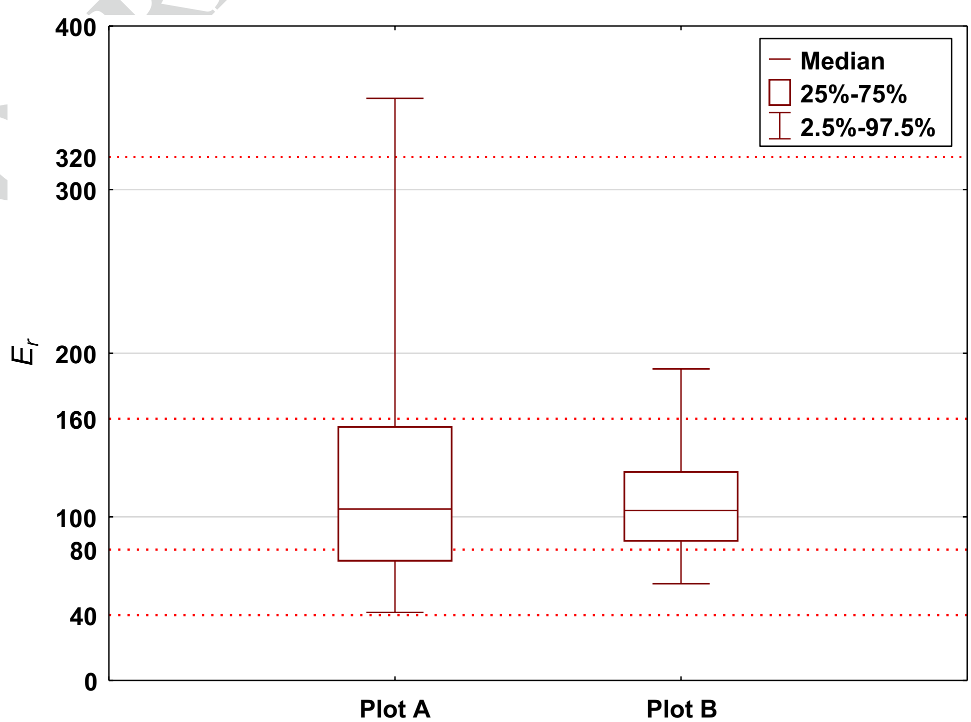

Fig. 3 Box-whisker plots of the ecological risk index (E_r) of soil As levels in the studied sites. The red line of dashes was the upper limit of low risk ($E_r = 40$), moderate risk ($E_r = 80$), considerable risk ($E_r = 160$), and high risk ($E_r = 320$), respectively

and 8.31% of the studied soil exhibited high risk in plot A and plot B, respectively, with E_r values ranging between 160 and 320. Additionally, approximately 3.48% of the soil samples in plot A were categorized as very high risk ($Er > 320$) according to the ecological risk categories previously illustrated. The higher variability in Er value in plot A may be due mainly to the higher variability of soil As concentration in this plot.

Elevated As levels in farmland soils may affect normal growth and reduce the yield of crop plants, including rice. It was reported that increased levels of As can induce straighthead disease, adversely affect photosynthesis, and negatively affect the growth parameters of rice (Kumarathilaka et al. 2018). Rice is a major cereal crop, and As contamination in rice paddy fields is a growing problem (Mridha et al. 2022). Generally, the background total As level in soil ranges from 5 to 6 mg kg^{-1}; however, the repeated application of As-containing pesticides and fertilizers has resulted in an increase in As accumulation level in farmland soils (Juang et al. 2021). For instance, it was reported that the total As level in agricultural soils in many countries, including Taiwan, exceeded the permissible limit (20 mg kg^{-1}) established by the US EPA (Azam et al. 2016; US EPA 2002). In this study, the ecological risk index (E_r), taking into consideration both the background level and biological toxicity of As, was adopted to evaluate the impact of As on the agricultural ecosystem. The mean As concentrations in both plots were higher than the background As level of local soils (i.e., 5.65 mg kg^{-1}), thus indicating considerable ecological risk. Interestingly, the E_r values were nearly the same for both plots, although the mean As concentration in plot A (70.94 mg kg^{-1}) was relatively higher than that of plot B (61.80 mg kg^{-1}). Furthermore, the E_r values obtained from the deterministic approach were higher than those from the probabilistic approach. This inconsistent result might be due mainly to uncertainties associated with these two plots (Zhang et al. 2021). Most previous studies conducting a phytotoxicity assessment of heavy metals have been based on deterministic analysis, which provided only limited information on heavy metal pollution and the underlying risk. Therefore, the probabilistic phytotoxicity assessment proposed in this study provides more detailed pollution information for better soil pollution management and control.

Daily iAs accumulation and health risk assessment

The parameters and input values used for health risk calculation are summarized in Table 3. The CTE of EDI in plot A was 9.5×10^{-5} and 7.1×10^{-5} mg kg^{-1} d^{-1} for local males and females, respectively, whereas the CTE of EDI

Table 3 Parameters and input values used in assessing human health risk of iAs associated with rice consumption

Parameter	Symbol	Input value	Unit	Source
Total As concentration in soil	C_{soil}		mg kg^{-1}	This study
Plot A		LN (49.07, 1.99)		
Plot B		LN (49.37, 1.44)		
Rhizosphere As fraction	f_{rhizo}		–	This study
Plot A		N (0.349, 0.058)		
Plot B		N (0.121, 0.010)		
Bioconcentration factor	BCF		–	This study
Plot A		LN (0.01183, 1.2509)		
Plot B		LN (0.04867, 1.2076)		
Ingestion rate	IR		g day^{-1}	FDA (2022)
Male		LN (109.18, 1.82)		
Female		LN (63.87, 1.87)		
Proportion of iAs of total As in brown rice	P_{inorg}	0.6836	–	This study
Proportion of iAs of white rice to that in brown rice	$P_{white/brown}$	0.65	–	Sun et al. (2008); Naito et al. (2015); Narukawa et al. (2011)
Bioaccessibility	BAc	Be (4.91, 1.85)	–	Zhou et al. (2021)
Conversion factor	CF	10^{-3}	kg g^{-1}	–
Body weight	BW		kg	MHW (2022)
Male		N (75.4, 21.6)		
Female		N (58.7, 14.7)		
Reference dose	RfD	0.0003	mg kg^{-1} d^{-1}	IRIS database
Slope factor	SF	1.5	kg d mg^{-1}	IRIS database

in plot B was 1.4×10^{-4} and 1.0×10^{-4} mg kg^{-1} d^{-1} for local males and females (Fig. 4(A)). A large variation in EDI was observed for males compared to females; thus, the RME of EDI was found to be 7.4×10^{-4} mg kg^{-1} d^{-1} for males in both plots. According to the IRIS database provided by the US EPA, the *RfD* of iAs for noncarcinogenic effect is 3×10^{-4} mg kg^{-1} d^{-1}. Therefore, HQ was calculated by Eq. (5) and is represented in Fig. 4(B). As can be seen, the median HQs for all exposure scenarios were below 0.5, indicating low noncarcinogenic risk in the average situation. However, all the 97.5%-tile HQs exceeded unity, implying potential noncarcinogenic risks from a conservative viewpoint. As for the carcinogenic effect, the median TRs ranged from 1.1×10^{-4} (plot A, female) to 2.1×10^{-4} (plot B, male), whereas the 97.5%-tile TR ranged from 8.2×10^{-4} (plot A, female) to 1.1×10^{-3} (plot B, male) (Fig. 4(C)). Therefore, all the considered exposure scenarios would pose unacceptable carcinogenic risks to the target populations, whether from an average or a conservative perspective.

Dietary habit is an important factor in human health risk assessment. In most Asian countries, including Taiwan, white rice is the major subtype of rice consumed by inhabitants. A number of previous studies have reported that the proportion of iAs in rice husk and bran is higher than in endosperm (Wang et al. 2019; dos Santos et al. 2021). As a result, the iAs fraction in white (polished) rice is lower than in brown rice. International regulations thus set different acceptable limits for iAs in brown rice and white rice. For example, the European Food Safety Authority (EFSA) sets regulatory limits for the adult population at 0.20 mg kg^{-1} for white rice and 0.25 mg kg^{-1} for brown rice (Islam et al. 2016). In this study, the parameter $P_{\text{white/brown}}$ was introduced in Eq. (4) to represent the proportion of iAs in white rice compared to that in brown rice. This parameter is significantly influenced by the degree of polishing (DOP). Naito et al. (2015) indicated that inorganic As levels in white rice polished by removing 10% of the bran by weight was reduced to 51–70% of those in brown rice. Narukawa et al. (2011) analyzed rice samples collected from various regions in Japan and found that iAs levels in white rice declined to 60% after a 10% DOP of brown rice. From a survey of rice samples obtained from Japan and the US, Sun et al. (2008) reported that iAs concentrations in white rice decreased to 43–83% after 7% DOP. By taking into consideration the DOP in Taiwan, therefore, an input value of 0.65 was adopted for $P_{\text{white/brown}}$ in this study.

The ingestion rate of white rice (IR) is another critical factor in oral iAs exposure and varies widely from country to country. It was reported that the per capita consumption of rice is highest at 400–650 g day^{-1} in Bangladesh, whereas rice consumption for a Brazilian adult is only 55 g day^{-1} (Islam et al. 2017; dos Santos et al. 2021). Additionally, significant gender differences have been observed. For instance, Ohno et al. (2007) conducted a survey in Bangladesh and reported that the intake of As is higher for males than females due to the greater daily rice consumption of males (776 g day^{-1}) compared with females (553 g day^{-1}). More recently, Chen et al. (2016) indicated that the daily intake of iAs varies considerably in Taiwan, mainly due to differences in the consumption rate of rice between males and females. In the present results, the estimated daily intake of iAs (EDI), as well as the subsequent non-cancer (HQ) and cancer risk (TR), were all higher for males than for females, mainly because the ingestion rate of white rice of males is greater than that of females. These findings are consistent with those published in previous studies (Ohno et al. 2007; Chen et al. 2016).

Apart from As-contaminated drinking water, it is generally recognized that rice consumption is the major route of As exposure in many Asian countries (Islam et al. 2016; Mridha et al. 2022). A number of studies have thus focused on As accumulation in rice and the subsequent human health risk. Conventionally, the risk assessment for dietary exposure to iAs is conducted based on the total iAs concentration in rice grain. However, Li et al. (2017) indicated that As intake based on total iAs in rice may overestimate As exposure 2.0- to 3.7-fold compared to that based on bioaccessible iAs. Therefore, risk assessments of iAs exposure from rice consumption should take bioaccessible iAs concentration (BAc) into consideration. The BAc of iAs depends mainly on the rice genotype and the preparation and cooking conditions of the rice (Yager et al. 2015). Recently, both in vivo animal models (e.g., swine or murine models) and in vitro digestion methods (e.g., physiologically based extraction test method, gastrointestinal method, unified BARGE method) have been developed for the determination of BAc (Laparra et al. 2005; Yager et al. 2015; Islam et al. 2017; Li et al. 2017; 2021; Wang et al. 2021). Large variations in the bioaccessibility of iAs in different rice genotypes have been reported in these studies. For example, Laparra et al. (2005) used a simulated gastrointestinal digestion method and found that the BAc of iAs in cooked rice ranged from 63 to 99%; however, Du et al. (2019) used a similar approach and reported that the average BAc of As (III) was only 55.1%. Furthermore, Li et al. (2017) indicated that the BAc of iAs in rice varies widely even within a country. As a result, Zhou et al. (2021) employed a beta distribution with parameters of $\alpha = 4.91$ and $\beta = 1.85$ to better characterize the uncertainty of BAc in rice. In this study, therefore, the distribution suggested by Zhou et al. was directly adopted to consider the uncertainty and variability of BAc in various rice genotypes. The resulting distribution of BAc ranged from 0.43 to 0.95 (5%-tile to 95%-tile), with a median of 0.75, which was comparable to the BAc values reported in previous studies (Laparra et al. 2005; Du et al. 2019).

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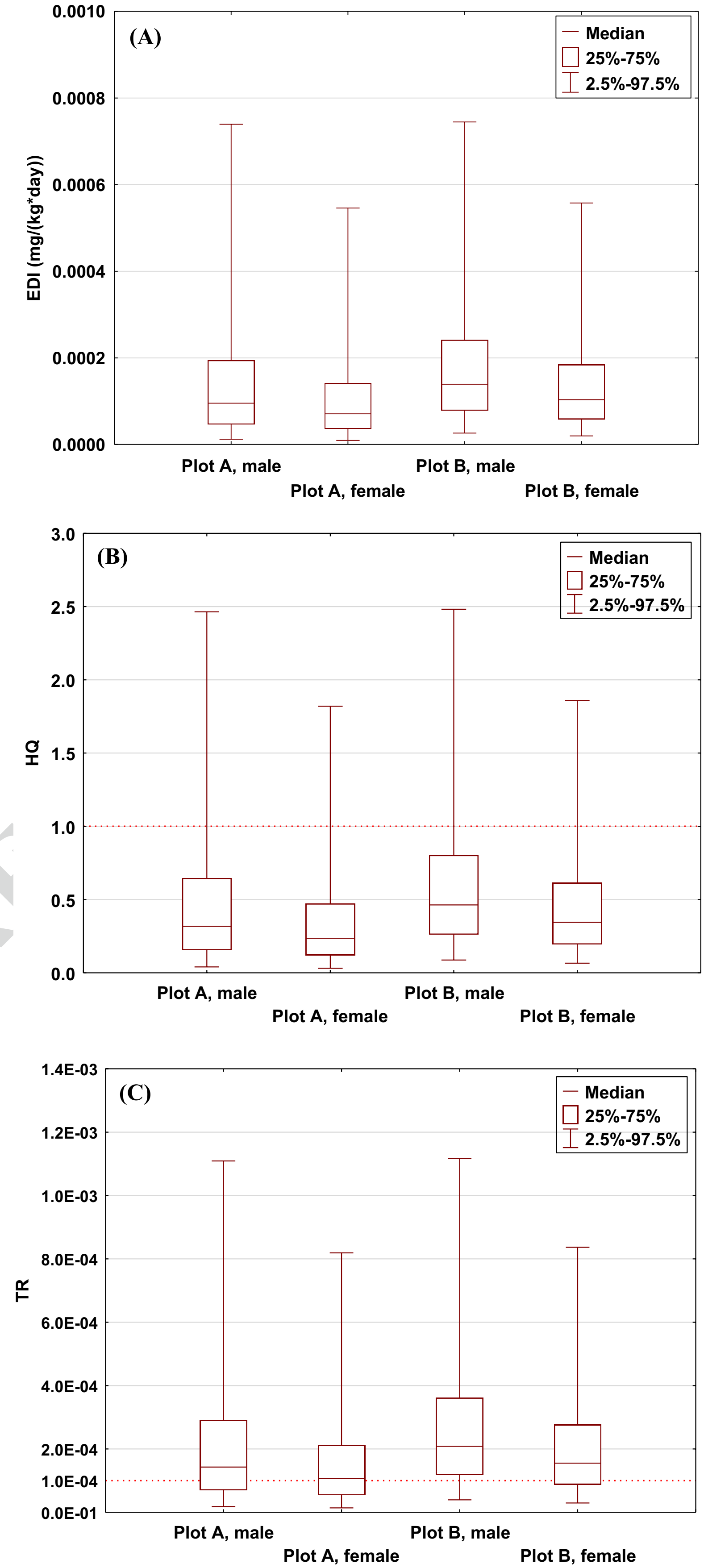

Fig. 4 Box-whisker plots of **A** estimated daily intake of iAs (EDI); **B** hazard quotient (HQ); **C** target cancer risk (TR) of residents (male and female) associated with consuming rice cultivated in the studied soils (plot A and plot B). The red line of dashes was the upper limit of acceptable noncarcinogenic risk (HQ = 1) and carcinogenic risk ($TR = 10^{-1}$) in **B** and **C**, respectively

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To characterize the effect of input parameters in Eq. (4) on health risk, sensitivity analyses were performed using Crystal Ball software. According to the results, the factors affecting health risk in plot A in decreasing order were as follows: $C_{soil} > IR > BW > BCF \approx BAc > f_{rhizo}$, whereas the factors affecting health risk in plot B in decreasing order were $IR > C_{soil} > BW > BAc > BCF > f_{rhizo}$ (Fig. 5). By definition, the sensitivity of a parameter represents how changing a unit in the value of the parameter changes the final result (Zhang et al. 2021). The inconsistency of the rank of the first two main factors (i.e., C_{soil} and IR) in the two plots may be due to the high variation of C_{soil} in plot A. Theoretically, based on the sensitivity analysis results, it will be most efficient to minimize the risk of local residents exposed to iAs via rice consumption by reducing the total As in soil or by reducing the ingestion rate of white rice. In reality, however, a reduction in rice ingestion is not an option in many parts of the world where rice is an irreplaceable part of the diet (Zhuang et al. 2016). Consequently, more effort should be put into the reduction of As levels in the soil. It has been confirmed that the main source of soil As is As-contaminated irrigation water (Majumder and Banik 2019). From the perspective of source control, therefore, priority should be given to the reduction of As concentrations in irrigation water. Recent studies have recommended that an electrocoagulation process be utilized for the removal or treatment of As in water (Sandoval et al. 2021). On the other hand, body weight (BW) is also a sensitive parameter. The body weight of males is higher than that of females. However, the effect of the higher ingestion rate of rice by males in the present study overwhelmed that of the greater body weight, so a larger EDI was found for males. For reducing BCF, the application of silicate materials in farming practice, as well as gene modification of rice cultivars have been proven to be promising and effective techniques to reduce As in rice grains (Juang et al. 2021). Although rhizosphere As fraction (f_{rhizo}) seems to be less sensitive, some researchers have suggested that the application of sulfur fertilizers and silicate materials can decrease the accumulation of As in rhizosphere or the mobility of soil As and thus reduce the health risk associated with rice consumption (Juang et al. 2021; Lü et al. 2022).

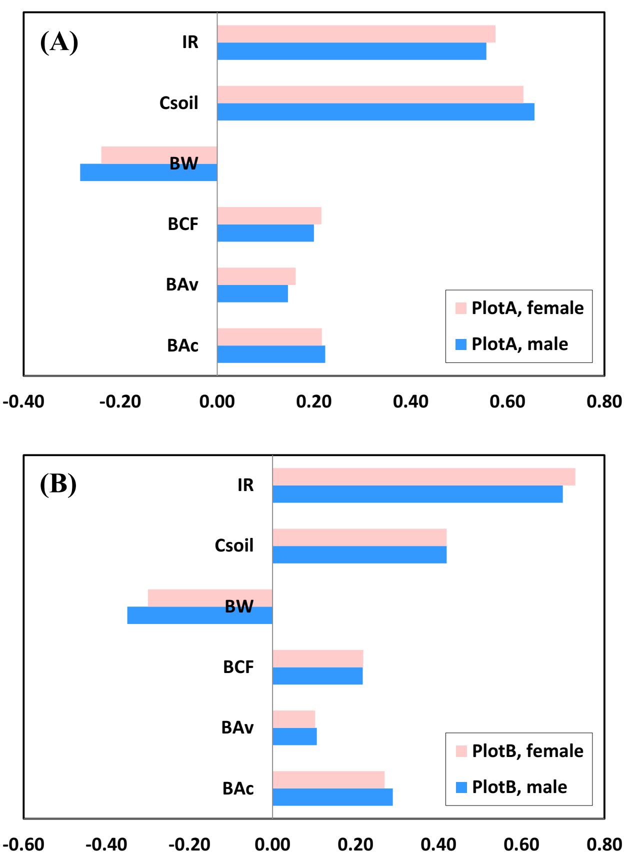

Fig. 5 Sensitivity analysis of health risk for local females and males associated with consuming rice cultivated in **A** plot A; **B** plot B

In order to examine the current soil quality standard for As, normal distributions with different mean values (i.e., 2.5, 5, 10, 15, 30, and 60 mg kg^{-1}) were considered various exposure scenarios and were employed to characterize C_{soil} in Eq. (4). The standard deviation was assumed to be 10% of its mean value. Then, the assumed distributions of C_{soil}, with all other parameters and input values in Table 3, were introduced into Eq. (4) to calculate the corresponding E_r, HQ, and TR. From a conservative perspective, the phytotoxicity of As in plot B, as well as the human health risk of males associated with the consumption of rice from plot B, were regarded as the worst exposure scenario and used for risk estimation. The simulation results are summarized in Table 4. At present, the soil quality standard for total As level in farmland soil is 60 mg kg^{-1} in Taiwan (EPA-TW 2011). According to Table 4, this standard will result in a "considerable" phytotoxicity and an "unacceptable" carcinogenic health risk even based on the CTE condition. In fact, the quality standard for As in farmland soil in Taiwan is higher than that in many other countries, such as Canada (12 mg kg^{-1}), the USA (20 mg kg^{-1}), and China (30 mg kg^{-1}) (Azam et al. 2016; Alexakis et al. 2021; Gao et al. 2021). On the other hand, when the mean value of C_{soil} is lower than 15 mg kg^{-1}, the phytotoxicity will become "low," and the noncarcinogenic risk will be acceptable even in the worst (i.e., RME) situation; however, the 97.5%-tile value of TR is still higher than 1×10^{-4}, indicating an unacceptable carcinogenic risk. The 97.5%-tile TR values were 1.8×10^{-4} and 9.3×10^{-5} when the mean C_{soil} values were decreased to 10 and 5 mg kg^{-1}, respectively. From the viewpoint of phytotoxicity and health risk assessment, therefore, the soil quality standard for As for two population groups consuming rice from different sampling sites is advised to fall

Table 4 Estimated ecological risk (*Er*), hazard quotient (*HQ*), and target cancer risk (*TR*) of iAs associated with consuming white rice according to soil As concentration (C_{soil})

C_{soil} (mg kg^{-1})	*Er*		HQ		TR	
	Median	97.5%-tile	Median	97.5%-tile	Median	97.5%-tile
60	106.1	128.8	0.56	2.49	2.5×10^{-4}	1.1×10^{-3}
30	52.8	64.1	0.28	1.21	1.3×10^{-4}	5.4×10^{-4}
15	26.5	32.2	0.14	0.63	6.3×10^{-5}	2.8×10^{-4}
10	17.6	21.5	0.09	0.41	4.2×10^{-5}	1.8×10^{-4}
5	8.8	10.7	0.05	0.21	2.1×10^{-5}	9.3×10^{-5}
2.5	4.4	5.4	0.02	0.10	1.1×10^{-5}	4.7×10^{-5}

within 5–10 mg kg^{-1}. However, other aspects, including risk communication, risk attitude, and risk management, should be also included by policymakers while setting the soil quality standard for As in farmlands.

In this study, limitations existed in the health risk assessment of As in rice. First, the influence of the cooking method on bioaccessible As was not considered, which would overestimate the health risk. In fact, rice is not consumed directly without cooking by nearly all consumers. Second, As intake through rice consumption was only one exposure pathway for local residents, which would underestimate the health risk. Indeed, it is generally recognized that other dietary exposure routes, such as drinking water and seafood, would also contribute a certain amount of daily As intake and pose a considerable health risk to human beings. To overcome the limitations of this study, therefore, further research work should be dedicated both to the effects of cooking on bioaccessible As and to the diet structure of residents.

Conclusions

The results indicated that a significant variation in the BCF value of As existed among different rice genotypes, and a negative correlation was observed between BCF value and rhizosphere As level in the soil. Second, the soil As level in the studied plots would result in considerable phytotoxicity on rice. Third, based on a conservative perspective, local residents would be exposed to unacceptable carcinogenic and noncarcinogenic health risks associated with consuming rice grown in the studied sites. Sensitivity analysis results further implied that priority should be given to reducing As levels in soils as well as in irrigation waters. Lastly, it was recommended that, for the protection of farmland ecosystems and human health, the current soil quality standard for As in farmlands should be decreased from 60 to 5–10 mg kg^{-1}. The methodology developed in this study could also be applied to provide the basis for refining and revising the soil quality standard for heavy metals in farmland in other regions and countries.

Author contribution All authors contributed to the study's conception and design. Conceptualization: Kai-Wei Juang and Bo-Ching Chen; methodology: Bo-Ching Chen; formal analysis: Bo-Ching Chen; investigation: Kai-Wei Juang, Li-Jia Chu, and Chien-Hui Syu; resources and data curation: Li-Jia Chu and Chien-Hui Syu; software: Kai-Wei Juang and Li-Jia Chu; writing—original draft preparation: Bo-Ching Chen; writing—review and editing: Bo-Ching Chen; supervision and funding acquisition: Bo-Ching Chen.

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Data availability The data used and/or analyzed during the current study are available from the corresponding author upon reasonable request.

Materials availability The data used and/or analyzed during the current study are available from the corresponding author upon reasonable request.

Declarations

Ethical approval The authors confirm that the Committee on Publication Ethics (COPE) guidelines have been adhered to in the submission of this manuscript.

Consent to participate Not applicable.

Consent for publication The authors all agree to the publication of this journal. No other consents for publication were required.

Competing interests The authors declare no competing interests.

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