Journal of Hazardous Materials 286 (2015) 179–186

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Arsenic accumulation and speciation in rice grains influenced by arsenic phytotoxicity and rice genotypes grown in arsenic-elevated paddy soils

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HIGHLIGHTS

- As concentrations in grains of different rice genotypes were reduced by As toxicity.
- Indica have equal or higher As concentrations than japonica in grains in this study.
- The concentrations of DMA, instead of As^{III}, increased with total As in rice grains.

ARTICLE INFO

Article history:
Received 27 September 2014
Received in revised form 9 December 2014
Accepted 27 December 2014

Keywords:
Arsenic
Arsenic species
Rice
Genotypes
Phytotoxicity
Paddy soil

ABSTRACT

Rice consumption is a major route of As exposure to human for the population of worldwide. This study investigates the effect of phytotoxicity and rice genotypes on the content and speciation of As in rice grains grown in different levels of As-elevated paddy soils from Taiwan. Three levels of As-elevated soils and six rice genotypes commonly planted in Taiwan were used for this study. The results indicate that As contents in grains of rice is not proportional to soil As concentrations and they were equal or higher in indica genotypes than japonica genotypes used in this study. It was also found that the As phytotoxicity not only reducing the grain yields but also the As concentrations in grain of rice. The predominant As species found in rice grains were dimethylarsinic acid (DMA) and arsenite. The concentrations of DMA increased with total As concentrations, wherggeas the arsenite remained in a narrow range from 0.1 to 0.3 mg kg^{-1}. Because of the lower toxicity of DMA than inorganic As species, the health risks may not be increased through consumption of rice even when total As content in the grains is increased.

1. Introduction

Inorganic arsenic (As) is identified as a non-threshold, class 1 human carcinogen, and the intake of As through rice consumption may lead to serious health effects such as bladder and skin cancers [1,2]. The sources of As in rice paddy fields include natural (biogeochemical process) and anthropogenic (As-containing irrigation water, metal mining activity, arsenical pesticides and fertilizer applications) pathways [3,4]. Rice is the dietary staple for about half of the world's population and unfortunately, rice consumption has been the main arsenic exposure route in recent years [1,5]. This is due to the high bioavailability and mobility of As in flooding conditions, enhancing the uptake and accumulation of As by rice plants [6]. The concentration of As accumulated in rice grains is approximately 10-fold higher than other cereal crops [5,7]. Meharg and Rahman [8] found that the concentrations of As in rice grains grown in As-contaminated soils in certain parts of Bangladesh to be up to 1.8 mg kg^{-1}, resulting in serious As related risks to the local residents. Moreover, paddy rice grown in As-contaminated soils result in As phytotoxicity (inhibition of ATP formation and oxidative stress) which causes lower grain yields [9,10].

In pore water found in paddy soil, the concentration and speciation of As are controlled by the soil redox status and other soil properties such as soil pH, organic matter, aqueous chemistry [the concentration of phosphorus (P) and silicon (Si)], clay minerals and iron oxides content [4,11–13]. In submerged soils, the mobility of arsenite (iAs^{III}; as H_3AsO_3) is higher than arsenate (iAs^{V}; H_3AsO_4) because of the reductive dissolution of iron (Fe) oxides/hydroxides

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http://dx.doi.org/10.1016/j.jhazmat.2014.12.052
0304-3894/

Table 1
Total As concentrations in grains of rice grown in different countries.

Country	Grain As (mg kg^{-1})	n	Reference
Bangladesh	0.05–2.05	35	Islam et al. [48]
China	0.02–0.46	124	Meharg et al. [23]
India	0.18–0.31	133	Meharg et al. [23]
Italy	0.07–0.33	38	Meharg et al. [23]
Spain	0.05–0.82	76	Meharg et al. [23]
Taiwan	0.10–0.63	280	Lin et al. [49]
Thailand	0.01–0.39	54	Meharg et al. [23]
USA	0.10–0.66	134	Williams et al. [5]
Vietnam	0.03–0.47	31	Phuong et al. [50]

and consequent reduction of iAs^{V} to iAs^{III} [14]. The uptake and translocation of As in rice plants and the accumulation in rice grains are strongly dependent on the As species that exist in the rhizosphere [15]. In addition, the presence of P and Si in soils also impact the uptake of As by paddy rice [16–18]. There are two main mechanisms involved in As uptake into the roots of rice plants. The first route is through the phosphate transport pathway since arsenate is an analogue of phosphate [19]. The second route is when arsenite (silicic acid analog) and undissociated methylated As species (dimethylarsinic acid (DMA) and monomethylarsonic acid (MMA)) are taken up into the root by aquaporin channels [20,21]. Raab et al. [22] indicated that the uptake efficiency of methylated As species (DMA and MMA) into the root was much lower than inorganic arsenic species (iAs^{III} and iAs^{V}), but the translocation efficiency in the rice plant of methylated As species was much higher than inorganic arsenic species. It is generally believed that the toxicity of inorganic As is much higher than the pentavalent methylated As species [23,24]. Since the As species distribution in rice grains governs human toxicity, therefore, understanding the As species in rice grains is important for evaluating the As toxicity of rice to humans. In our previous studies, we found that the translocation of As in rice plants impacts on the accumulations of As in rice shoots among different rice genotypes in As-contaminated soils [25]. In this study, we intend to further investigate the effects of rice genotypes on As accumulation and speciation in rice grains grown in As-elevated paddy soils from the Guandu Plain of northern Taiwan.

In general, DMA and iAs are the predominant As species in rice grains [26] and their percentages varied widely. Many studies have shown the effect of rice genotypes and environmental factors on the accumulation and speciation of As in rice grains [27–29]. Table 1 shows total As concentrations in grains of rice grown in different countries. It indicated that the maximum As concentrations in grains differed by 6–7 folds in rice grown in different countries. It also discovered that grain As concentrations differed by about 40-folds in rice produced in the same country, indicating that the genotypes and environmental factors play an important role in grain As accumulation. Norton et al. [28] and Ahmed et al. [27] reported that the environment was the main controlling factor in grain As concentrations. Norton et al. [29], and Pillai et al. [30] indicated that there were variation in grain As concentrations among different cultivars, and there were significant genotype effects on the As speciation in rice grains. In addition, As phytotoxicity also impacts on the accumulation and speciation of As in rice grains [31]. Many studies have investigated rice As uptake and accumulation in soils of As concentrations less than 100 mg kg^{-1} [27–30,32]. Norton et al. [28] evaluated the accumulation of As in grains of rice grown at two field sites in Bangladesh (soil As: 10.3 ± 2.2 and 29.6 ± 7.2 mg kg^{-1}), India (soil As: 6.3 ± 1.3 and 17.9 ± 4.0 mg kg^{-1}) and China (soil As: 64.6 ± 4.7 and 65.6 ± 2.5 mg kg^{-1}), respectively. In addition, Hsu et al. [32] also investigated As concentrations in grains of rice grown in paddy soils in southwestern Taiwan (soil As: 52.2 ± 27.1 mg kg^{-1}). However, There are only few studies that have investigated rice As uptake and accumulation in high level of As-contaminated soils (> 200 mg As kg^{-1}). The As accumulation and speciation in rice grain among different rice genotypes grown in different levels of As-contaminated soils is worthy to be studied. Therefore, the aim of this study is to investigate the effect of As phytotoxicity and rice genotypes on the content and speciation of As in rice grains.

Table 2
The basic properties of the three levels of As-contaminated soils of Guandu Plain.

	pH	Org. C(%)	Texture	Fe_d	Al_d	Fe_o	Al_o	As_o	As_{total}
Test soils				g kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}	mg kg^{-1}	mg kg^{-1}
As-L	6.6	3.6	Clay	20.0	3.7	10.5	1.9	8.3	16.3
As-M	5.1	3.6	Clay	21.9	2.6	21.7	2.2	257.4	343.3
As-H	4.9	2.1	Clay	36.6	4.3	25.3	3.0	334.1	512.3

Fe_d dithionite–citrate–bicarbonate extractable Fe; Al_d dithionite–citrate–bicarbonate extractable Al; Fe_o ammonium oxalate extractable Fe; Al_o ammonium oxalate extractable Al; As_o ammonium oxalate extractable As.

2. Material and methods

2.1. Soil collection and characterization

Three levels of As-contaminated soils (16.3 (As-L), 343.3 (As-M) and 512.3 (As-H) mg As kg^{-1}) used in this study were collected from the surface soil (0–30 cm) of paddy fields in the Guandu Plain, Taipei, Taiwan (Table 2). The As concentrations of As-M and As-H soils are higher than farmland control standard (60 mg kg^{-1}) of Taiwan's Environment Protection Administration. Basic properties of tested soil are presented in Table 2.

2.2. Pot experiment of paddy rice

Pot experiments were performed in the phytotron at controlled temperature (20/25 °C, night/day) and relative humidity (70–95%) under sunlight. Six rice genotypes commonly planted in Taiwan including three japonica rice (TK 9, TC 192, TK 139) and three indica rice (TCN 1, TCSW 1, TCSY 837) were used in this study. There were three replicates for each rice genotype grown in three levels of As-contaminated soils. Rice seeds were sterilized in a solution containing 1% sodium hypochlorite (NaClO) solution and 1 drop of Tween 20 for 30 min, washed with deionized water and then germinated in Petri dishes containing moist tissue paper for three days. After germination, rice seedlings were transferred to a 0.6-L beaker and grown in half-strength modified Kimura B nutrient solution (pH was adjusted to 4.8–5.0 and the solution was renewed every two days) for 2 weeks. Two seedlings were transplanted into each pot filled with 2.3 kilograms of tested soil. The soils were saturated with water and the water level was maintained at about 3–5 cm above the soil surface during the whole period of plant growth. The soils were supplemented with 0.26 g N kg^{-1} as $NH_4{}_2SO_4$, 0.039 g P_2O_5 kg^{-1} as KH_2PO_4 and 0.054 g K_2O kg^{-1} as K_2SO_4 as basal fertilizers. The application of 0.13 g N kg^{-1} as a top dressing was done 15 days and 60 days after transplantation respectively. Mature rice was harvested nearly 130 days after transplantation. The harvested rice plants were separated into grain, flag leaf, straw and root. These samples were rinsed with tap water and then with deionized water. In order to avoid changes of As species in plant tissues, a portion of plant samples were stored at −20 °C till As species analysis. The grain yield, biomass of rice plants and lengths of each shoot and root were measured.

2.3. Soil pore water collection and analysis

Soil pore water in the pots was collected by Rhizon soil moisture samplers (Rhizosphere Research Products) inserted into the