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Increase of As release and phytotoxicity to rice seedlings in As-contaminated paddy soils by Si fertilizer application

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HIGHLIGHTS

- Si fertilization increases As release in As-contaminated paddy soils.
- Si fertilization into As-contaminated paddy soils could enhance As phytotoxicity.
- Decrease of As uptake by paddy rice was not observed by Si fertilization.

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ABSTRACT

Silicon (Si) was shown to be able to reduce arsenic (As) uptake by rice in hydroponic culture or in low As soils using high Si application rates. However, the effect of Si application on As uptake of rice grown in As-contaminated soils using Si fertilizer recommendation rate has not been investigated. In this study, the effect of Si application using Si fertilizer recommendation rate on As release and phytotoxicity in soils with different properties and contents of As was examined. The results show that the concentrations of As in soil solutions increased after Si applications due to competitive adsorption between As and Si on soil solids and the Si concentrations in soil solutions were also elevated to beneficial levels for rice growth. The rice seedlings accumulated more As and its growth was inhibited by Si application in As contaminated/spiked soils. The results indicate that there is an initial aggravation in As toxicity before the beneficial effects of Si fertilizing to rice were revealed when Si application based on fertilizer recommendation rate to As-contaminated paddy soils. Therefore, for As-contaminated paddy soils with high levels of As, the application of Si fertilizer could result in increasing As phytotoxicity and uptake by rice.

1. Introduction

Although the concentration of Arsenic (As) in the environment is generally low, it is widely distributed and is known as a human carcinogenic metalloid. Therefore, investigating As mobility and bioavailability in soils has become an important issue. In anaerobic soil environments, arsenite is the major species and its proportion among As species in soil solutions of paddy field is high [1,2]. Paddy rice is a staple foodstuff in Asia and unfortunately it accumulates As more efficiently in comparison with other cereals [3]. Excessive As accumulation in the rice grain can pose a significant health risk to consumers [4–6]. Moreover, the phytotoxicity of As is increased by a high level of As content in soils and consequently the rice growth or yields in paddy fields decrease [7–9].

Paddy rice requires higher amounts of Si than other plants [10]. Si can protect rice from diseases by physically strengthening cell walls to form a physical barrier as a host defense mechanism and also enhance the resistance of plants to biotic and abiotic stresses [11,12]. Therefore, Si fertilizer has been used to enhance the health and productivity of rice in areas with low Si availability via either soil or foliar applications. Moreover, the ability of Si to ameliorate metal toxicities has been increasingly studied in recent years [13–15]. Because arsenite and silicic acid both have a high pK_{a1} (9.0–9.3) and are tetrahedral with similar sizes, arsenite shares the Si transport pathways for entry into rice root cells [16–18]. Consequently, Si and As may compete against each other for the uptake and translocation in rice plants. In solution cultured experiments, adding Si to the nutrient solutions resulted in less As accumulation in rice plants [8,17,19,20].

However, since the behavior of Si is similar to that of As, Si can also compete against As for retention sites on soil surfaces, and thus the application of Si into soils might increase As concentration in soil solutions. Silicate was found to reduce arsenite adsorption rates and block potential adsorption sites thereby reducing the total quantity of arsentite adsorbed, and displace adsorbed arsenite [21]. The effects of Si addition to soils on arsenic uptake by rice are very complicated and both the enhancing and inhibition effects may occur as mentioned above.

A negative relationship between indigenous silicic acid in the soil solution and As uptake by rice in six uncontaminated soils was reported [22]. This implies that soils with high plant available Si contents resulted in low plant As contents. In a pot experiment, application of 20 g SiO_2 kg^{-1} (about 40 tons ha^{-1}) into As-uncontaminated soil decreased the total As concentration in straw and grain of rice by 78 and 16%, respectively [23]. The inhibition effect of As uptake by rice resulted from large amount of Si applications was reported in several studies [24–27]. In these

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experiments, the decrease in As uptake by rice was found when the As contents in the soils were less than 20 mg kg^{-1} and the Si application rates were high (about 20–150 tons SiO_2 ha^{-1}). However, the effect of Si application on As uptake by rice in As-contaminated soils containing high level of As or using low levels of Si based on Si fertilizer recommendation rate has not been investigated. Furthermore, the effects of soil properties which may control As–Si interactions in soil–rice systems on As uptake by rice as influenced by Si application are not clear. The objective of this study is to investigate the effects of different Si application rates on As release into soil solutions and on As toxicity to rice seedlings in soils with different properties and contents of As.

2. Materials and methods

2.1. Collection and As treatments of soils

Guandu soils were collected from the paddy fields of Guandu plain, northern Taiwan, which were identified as an As-contaminated area caused by irrigating with As-contaminated spring water [28]. Two Guandu (Gd) topsoils were used which have low (background; $Gd_{\text{-L}}$) and high (contaminated; $Gd_{\text{-H}}$) levels of As and both are still cultivated for rice production to date. Another four uncontaminated paddy soils were collected from the topsoils of Pinchen (Pc), Tainan (Tn), Neipu (Np) and Chiwulan (Ca) soil series which have low available Si contents and are widely distributed in Taiwan. The soils were air-dried, sieved through a 10-mesh sieve, well mixed and then stored in plastic containers (un-spiked $Pc_{\text{-L}}$, $Tn_{\text{-L}}$, $Np_{\text{-L}}$, and $Ca_{\text{-L}}$, respectively). A portion of Pc, Tn, Np, and Ca soils were spiked using disodium hydrogen arsenate solution. Two hundred milliliters of 1000 mg As L^{-1} solution was sprayed and mixed artificially with each of 1 kg soil within 10 min (i.e. 200 mg As kg^{-1} soil was added). The As-spiked soils ($Pc_{\text{-H}}$, $Tn_{\text{-H}}$, $Np_{\text{-H}}$, and $Ca_{\text{-H}}$) were then subjected to the cycling process of wetting (water-saturated) and drying followed by further sieving and mixing. Three cycling processes were done at room temperature for approximately 60 days of aging time. The spiking-aging procedure resulted in similar proportions of arsenate (20%) and arsenite (80%) to total As in soil solutions compared to those of the geogenically contaminated $Gd_{\text{-H}}$ soils. Subsamples of soils were taken and total As contents were analyzed. Accordingly, five As-uncomtaminated/unspiked soils ($_{\text{-L}}$) and five As-contaminated/spiked soils ($_{\text{-H}}$) of Gd, Pc, Tn, Np, and Ca were used. Some selected properties of the soils are given in Table 1.

2.2. Si treatments

In this study, the amounts of Si application is based on the Si fertilizer recommendation rate which is much lower than those used in the previous studies on As–Si competition for rice uptake mentioned above. According to a study by the Council of Agriculture, Taiwan, the application of silicate slag at the rate of 3 tons ha^{-1} (about 750 kg ha^{-1} 0.5 N HCl soluble Si) is recommended for rice cultivation in soils containing available Si lower than 40 mg SiO_2 kg^{-1}. The recommended rate of available Si approximately equals to 0.375 g Si kg^{-1} and was used in this study. We conducted a series of Si treatments by adding sodium silicate solutions into soils at the rates of 0, 1/4, 1/2, 1, and 2 fold of the recommended rate (denoted by −Si, +1/4Si, +1/2Si, +Si, and +2Si, respectively). In soil incubation and pot experiments, the Na^+ contents in soils were adjusted to the same level by adding NaCl solutions to compensate the difference caused by As-spiking or Si application. Soils were then applied with urea, monocalcium phosphate, and potassium chloride at the recommended rates of 110 kg N ha^{-1}, 100 kg P_2O_5 ha^{-1}, and 180 kg K_2O ha^{-1} as base fertilizers, respectively.

2.3. Soil incubation experiments

The 20 g of soil samples after As or Si treatments were added into 50 mL centrifuge tubes. The water contents were adjusted to reach 1:1 (soil:water weight basis) at the beginning and every 3 days after. The soils were then incubated at 25 °C in darkness for 0, 3, 6, 12, 24, 36 and 50 days, respectively. On the 27th day, 110 kg N ha^{-1} as urea was applied as the additional fertilizer. At the end of each incubation period, the moisture was adjusted to 1:1 soil/water ratio again and the centrifuge tubes were shaken at 180 rpm for 30 min followed by centrifugation at 10,000 rpm for 10 min. The supernatant was then passed through a 0.45-μm filter. The pH values of filtrates were measured and the concentrations of As and Si were analyzed after adequate dilution. Another set of 500 g of treated soils were incubated in 1 L plastic pots with the same conditions as the soil incubation experiments described above and the redox potential (Eh) was measured during the 50 days of incubation.

2.4. Pot experiments of rice seedlings

The cultivar of paddy rice (*Oryza sative* L.), Taichung 65, was used in this study because of its higher efficiency in Si accumulation among 47 cultivars commonly cultivated in Taiwan [29]. The details of seed germination and seedling raising procedures were same as those described in our previous research [30]. Five-hundred grams of soils treated with Si, fertilized, and followed by moisture adjustment as same as those in incubation experiments were placed in pots. The pots were then moved into a phytotron glasshouse with a controlled condition (day 25/night 20 °C; RH 70–95%) for 12 days. After that, 10 seedlings at the three-leaf stage were transplanted into each pot and additional N fertilizers were added on day 27. The rice seedlings were harvested at 38 days after transplanting. The biomass and the concentrations of As, Fe, and P of shoots and roots were measured.

2.5. Soil and plant analyses

The pH and Eh of soils before and during the incubation were measured using a glass electrode and platinum electrode, respectively. The soil organic carbon content was determined by the Walkley–Black wet combustion method [31]. The soil particle size distribution was measured by a hydrometer method [32]. The amorphous Fe content in soils was extracted by oxalate [33] while the available Si in soils was extracted by sodium acetate [34]. The soils were digested with hydrogen peroxide following the procedure adapted from Taiwan EPA [35] for determining total As concentration. The plant tissues were digested with HNO_3/H_2O_2 following the procedure adapted by Cai et al. [36] for determination of As and Si contents. The As concentrations in soil solutions of the incubation experiments and digested solutions of soils and plant tissues of background soils (−As) were determined using ICP-MS (Agilent, 7700x). While those in As-spiked/contaminated soils (+As) and the concentrations of other elements in soils and plant samples were determined by ICP-OES (Perkin-Elmer, Optima 2000). The statistical analysis was carried out by using the general linear model of SAS to test the difference in the measurements between treatments with Ducan's multiple range test (DMRT) at a level of $P = 0.05$ (SAS Institute).

3. Results and discussion

3.1. Effects of Si application on Si concentrations in soil solutions

Fig. 1 shows the change of Si concentrations in soil solutions during incubation for Si treatments (only results of control and +Si treatment are shown). It indicates that Si concentrations in all the tested soils were kept low during the incubation without Si application and that Si concentrations in soil solutions significantly increased after Si application ($P < 0.05$). The final Si concentration (50 days) in the solution of Si treated $Np_{\text{-L}}$ soil was 97 mg L^{-1}