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The availabilities of arsenic and cadmium in rice paddy fields from a mining area: The role of soil extractable and plant silicon*



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ABSTRACT

Adequate silicon (Si) can greatly boost rice yield and improve grain quality through alleviating stresses associated with heavy metals and metalloids such as arsenic (As) and cadmium (Cd). The soil plantavailable Si is relatively low in South China due to severe desilicification and allitization of the soils in this region. Conversely, pollution of heavy metals and metalloids in the soils of this region occurs widely, especially As and Cd pollution in paddy soil. Therefore, evaluating the plant availability of Si in paddy soil of South China and examining its correlation with the availability of heavy metals and metalloids are of great significance. Accordingly, in our study, 107 pairs of soil and rice plant samples were collected from paddy fields contaminated by As and Cd in South China. Significantly positive correlations between Si in rice plants and Si fractions in soils extracted with citric acid, NaOAc-HOAc buffer, and oxalate-ammonium oxalate buffer suggest that these extractants are more suitable for use in extracting plant-available Si in the soils of our present study. Significantly negative correlations between different Si fractions and As or Cd in rice plant tissues and negative exponential correlations between the molar ratios of Si to As/Cd in rice roots, straws, husks or grains and As/Cd in rice grains indicate that Si can significantly alleviate the accumulation of As/Cd from soils to the rice plants. Finally, a contribution assessment of soil properties to As/Cd accumulation in rice grains based on random forest showed that in addition to Si concentrations in soil or rice plants, other factors such as Fe fractions and total phosphorus also contributed largely to As/ Cd accumulation in rice grains. Overall, Si exhibited its unique role in mitigating As or Cd stress in rice, and our study results provide strong field evidence for this role.

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1. Introduction

Due to rapid urbanization, mining, metal processing, and applications of fertilizers and sewage sludge, the pollution of heavy metals and metalloids in soils occurs widely, especially arsenic (As) and cadmium (Cd) pollution in paddy soils (Yamaguchi et al., 2011; Wang et al., 2014). As a result, rice grown in these contaminated paddy soils is seriously polluted by As or Cd. On the other hand, rice is the staple food for approximately half of the world's population, especially for Asian people (Stone, 2008). Therefore, exploring

* Corresponding author. E-mail address: cefbli@soil.gd.cn (F. Li). effective measures for reducing the health risks to people posed by As or Cd via rice uptake is very urgent (Islam et al., 2014).

As the second-most abundant element in the Earth's crust, silicon (Si) is beneficial for rice growth, and it has been demonstrated to alleviate the deleterious effects of heavy metals and metalloids on rice grown in soils contaminated by As or Cd (Adrees et al., 2015; Meharg and Meharg, 2015). It was reported that adequate Si fertilization can greatly boost rice yield and improve grain quality through lowering the concentrations of As and Cd (Meharg and Meharg, 2015). Silicon fertilization of paddy soil reduces As uptake and translocation to plant shoots and grains, probably because Si, as silicic acid, outcompetes As (arsenite) for uptake (Meharg and Meharg, 2015). In contrast, the alleviation of Cd toxicity for Si is partially attributed to the enhancement of cell wall elasticity and plasticity by Si (Vaculik et al., 2009), as well as increased deposition

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of Cd in the cell walls (Vaculik et al., 2012). Although many previous studies regarding the effect of Si on the mitigation of heavy metal toxicities in plants have been reported, these studies mainly examined the mechanism of Si-induced mitigation of heavy metal toxicity in plants from the perspective of plant physiology or exogenous Si addition based on pot and hydroponic experiments (Wu et al., 2013; Rizwan et al., 2016), and supporting findings derived from in situ field experiments are rare.

Generally, Si in soils is predominantly in the form of silicates, quartz, biogenic SiO₂ (phytoliths and diatoms) and silica gel (Sommer et al., 2006). The extractable Si includes water-soluble Si, active Si and amorphous Si. Water-soluble Si is considered as a directly available form of Si for plants, whereas the active Si and amorphous Si can be transformed to water soluble Si under suitable conditions, and then they can be utilized by plants (Xiang et al., 1993). Because of weathering and leaching, desilicification and allitization of the soils located in the subtropical region are severe under hot and rainy weather conditions. Hence, the plant-available Si is relatively low, and Si-deficient paddy soils are widespread in South China (Cai, 2005). To evaluate soil plant-available Si concentration, a large number of single-extraction methods have been proposed based on the extraction capacities of different extractants, including citrate, acetate, oxalate, NaOH, Na₂CO₃ and tiron solutions (Biermans and Baert, 1977; Liu et al., 2006; Meunier et al., 2014). It is necessary to explore the methods suitable for assessing the concentration of the plant-available Si in the paddy soils of South China. Moreover, systemically investigating the correlations between Si fractions extracted with different extractants and the availabilities of heavy metals and metalloids from soil to plant is also very helpful for elucidating the mechanism of Si-induced mitigation of heavy metal toxicity in plants.

The Lianhuashan tungsten mine, which lies in Shantou City, Guangdong Province, is one of the largest tungsten mines in China. The paddy fields downstream of the mining area have received a substantial influx of As and Cd from acid mining drainage (Liu et al., 2015). The objective of this study is to assess the plant availabilities of different Si fractions and delineate the role of plant-available Si fractions in alleviating As and Cd stress on rice plants under field conditions. Hence, As and Cd concentrations in different parts of the rice plants and paddy soils collected from the study area were determined. Different Si fractions in soils were also determined using different extraction techniques.

2. Materials and methods

2.1. Sampling and soil properties

A total of 107 pairs of paddy soil and rice plant samples were collected from the downstream area of the Lianhuashan tungsten mine in November 2011. All the rice plants belong to Indica hybrid rice Y Liangyou based on the broad classification. At each sampling field, soil and rice plant samples were collected with a random approach, i.e., 3-5 sampling points for a field were set according to the sampling area, and the soil and rice plant samples taken from 3 to 5 sampling points were mixed as composite samples, respectively. All of the samples were sealed in polytetrafluoroethylene (PTFE) bags and then transported to the laboratory. In the laboratory, soils were uniformly air-dried and sieved through a 100 mesh stainless-steel screen for subsequent analysis. The harvested rice plants were separated into brown rice, husk, straw and root subsamples. These samples were rinsed with 0.1 M HCl, tap water and then with deionized water more than three times. After being dried at 60 °C in an oven for 72 h, rice plants were ground into fine powders (at least 0.05 mm particle size) either by hand using a Sifree mortar and pestle or with a motorized, stainless steel grinder (model BCG100WH1; Kitchen Aid, St. Joseph, Mich.) for further analysis. Sub-samples of soil were used to measure the soil properties including pH, total and available soil phosphorus (P), organic matter (OM), and cation exchange capacity (CEC), as well as soil Fe fractions including DCB-extractable Fe (Fe_{DCB}), amorphous Fe (Fe_O), complex Fe (Fe_C), HCl-extractable Fe(II) and Fe(III). The methods used for these analyses were described in a previous study (Yu et al., 2016).

2.2. Extraction methods for the silicon fraction in soil

The Si fraction of the soils was extracted using six, short-term, single extraction techniques, which were widely employed to evaluate plant-available Si in previous studies (Sauer et al., 2006). The first four extraction methods employed 0.19 M Na₂CO₃ (Na₂CO₃—Si) (pH = 11.8), 0.5 M NaHCO₃ (NaHCO₃—Si) (pH = 8.5), 0.025 M citric acid (CA-Si), and oxalate-ammonium and oxalate buffer (Ox-Si). A 5-g aliquot of air-dried soil was extracted with a 50-mL aliquot of extractant in a 100-mL polyethylene Erlenmeyer flask. The extraction was performed in a shaking tube at 40 °C and 120 revolutions per minute (rpm) for 5 h prior to filtration through Whatman No. 5 filter paper.

The fifth extraction method employed a NaOAc-HAc buffer (HAc) (pH = 4.0). A 5-g aliquot of air-dried soil was extracted with a 50-mL aliquot of extractant in a 100-mL polyethylene Erlenmeyer flask. The flask was placed in a 40 °C water bath (Orbit Water Bath Shaker, Cat. No. 3535, Lab-Line Instruments, Inc., Melrose Park, I L) for 5 h (Savant et al., 1997; Korndorfer et al., 2001). The sixth extraction method employed a DCB solution. A 1.0-g aliquot of airdried soil was added to 50-ml polyethylene centrifuge tubes and treated in a 80 °C water bath and shaken at 120 rpm for 15 min with DCB solution. Then, the mixture was centrifuged at 4000 rpm for 5 min, and the supernatant was collected into a 50 mL volumetric flask. The residue was treated with DCB solution once again, and the supernatant was collected into the same 50 mL volumetric flask. After that, the residue was treated with 5 mL 1 M NaCl to remove the citrate-dithionite reagent and centrifuged. The supernatant was collected into the same 50 mL volumetric flask, and then the volumetric flask was filled with deionized water for further analysis (Weaver et al., 1968).

2.3. Determination of silicon in soil

The Si concentrations of the first four extraction methods were determined by colorimetric analysis. The filtrates were refrigerated at 4 °C for no more than four days prior to colorimetric analysis. Soil-test Si levels were determined using a modified blue silicomolybdous acid procedure (4500-SiO₂ E) that employed 20% (w/w) tartaric acid instead of oxalic acid, and color absorption was measured at 660 nm (Xu et al., 2001). The Si concentrations extracted by the oxalate-ammonium and oxalate buffering and the DCB solution were determined with ICP-OES analysis (Georgiadis et al., 2013).

2.4. Plant silicon analysis

Silicon in plant tissue was determined by the autoclave-induced digestion method recommend by Elliot and Snyder (Elliott and Snyder, 1991). Briefly, 0.1 g of plant powder was placed into a 100 mL polyethylene tube, and 3 mL of 50% (w/w) NaOH was added and sealed with a loose-fitting plastic cap. After gently mixing on vortex, the mixture was autoclaved at 121 °C for 20 min, transferred to a volumetric flask and adjusted to 50 mL with deionized H₂O. For determination, 1 mL of sample solution was transferred to a 50 mL volumetric flask, and 30 mL of 20% (w/w) acetic acid and 10 mL

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