



Cadmium availability in rice paddy fields from a mining area: The effects of soil properties highlighting iron fractions and pH value



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ABSTRACT

Cadmium (Cd) availability can be significantly affected by soil properties. The effect of pH value on Cd availability has been confirmed. Paddy soils in South China generally contain high contents of iron (Fe). Thus, it is hypothesized that Fe fractions, in addition to pH value, may play an important role in the Cd bioavailability in paddy soil and this requires further investigation. In this study, 73 paired soil and rice plant samples were collected from paddy fields those were contaminated by acid mine drainage containing Cd. The contents of Fe in the amorphous and DCB-extractable Fe oxides were significantly and negatively correlated with the Cd content in rice grain or straw (excluding DCB-extractable Fe vs Cd in straw). In addition, the concentration of HCl-extractable Fe(II) derived from Fe(III) reduction was positively correlated with the Cd content in rice grain or straw. These results suggest that soil Fe redox could affect the availability of Cd in rice plant. Contribution assessment of soil properties to Cd accumulation in rice grain based on random forest (RF) and stochastic gradient boosting (SGB) showed that pH value should be the most important factor and the content of Fe in the amorphous Fe oxides should be the second most important factor in affecting Cd content in rice grain. Overall, compared with the studies from temperate regions, such as Europe and northern China, Fe oxide exhibited its unique role in the bioavailability of Cd in the reddish paddy soil from our study area. The exploration of practical remediation strategies for Cd from the perspective of Fe oxide may be promising.

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1. Introduction

Cadmium (Cd) contamination in rice has attracted much attention in Asian countries because it poses a serious threat to human health. As previously reported, Cd intoxication can severely damage several human organs and systems, such as the respiratory system, kidneys, reproductive system and skeletal system (Godt et al., 2006). Rice is a staple food for most Asian populations, for which the dietary intake of rice is a major source of Cd to the human body (Tsukahara et al., 2003). Due to rapid urbanization, mining and metal processing, Cd pollution occurs widely in China, especially in paddy soil of South China (Wang et al., 2014b). In a previous study, Cd contents were reported to exceed the allowable Cd limit (0.2 mg kg⁻¹) specified by the Chinese food security

standards in more than 10% of rice grain samples across China (Zhen et al., 2008).

Previous studies, especially studies from temperate regions, such as Europe, have demonstrated that metal phytoavailability is affected by soil properties, such as the pH, organic matter (OM) content, clay content, cation exchange capacity (CEC) and available phosphorus (P) content (Brown et al., 2004; Romkens et al., 2011; Pietrzykowski et al., 2014). Of these properties, pH significantly influences metal phytoavailability. A recent study (Rafiq et al., 2014) indicated that the Cd contents in rice grains were significantly and negatively correlated with soil pH. In addition, several previous reports have established multiple regression models based on the correlation between pH and the crop uptake of Cd (McBride, 2002; Rafiq et al., 2014). Generally, the mobility and bioavailability of Cd increase as the soil pH decreases, because lower pH values enhance the transformation of Cd from an immobile form (e.g., carbonate and Fe and manganese (Mn) oxide bound forms) to an easily

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bioavailable form (e.g., the exchangeable form) (Xian and Shokohifard, 1989; Li et al., 2014a). Compared with studies from temperate regions, soils in (sub) tropical areas feature an abundance of iron (Fe) oxides with a higher redox activity, especially in paddy soil (Li et al., 2006). Thus, we hypothesized that the bioavailability of Cd in paddy soil from this region is closely related to the Fe oxide fractions in addition to other soil physicochemical properties. However, these factors require further investigation. In fact, the transformations of Fe-containing minerals can significantly affect the mobility of Cd in the soil and increase or decrease the bioavailability of Cd. For example, Fe oxides possess a significant capacity for Cd adsorption and can effectively immobilize Cd (Liu et al., 2014b). The secondary Fe minerals produced by Fe(III)-reducing bacteria and the bacteriogenic Fe oxides produced by Fe(II)-oxidizing microorganisms can all immobilize Cd (Martinez et al., 2004; Muehe et al., 2013). In addition, the Fe(II)-catalyzed recrystallization of Fe-(hydro)oxides can result in the incorporation of Cd (Cooper et al., 2006). Moreover, the reduction of Cd-bearing Fe(III) minerals by Fe(III)-reducing bacteria can directly enhance the mobility of Cd (Muehe et al., 2013). However, these studies were mainly based on laboratory experiments. Studies regarding the effects of Fe redox cycling on the mobility of Cd in paddy fields in the red soil region remain rare. Furthermore, the chemical fractions of heavy metals can influence the bioavailability and mobility of metals (Santos et al., 2002). Particularly, sequential extraction procedures can be used to investigate the geochemical partitioning of heavy metals among solid mineral and organic phases. The chemical forms of Cd are classified as exchangeable forms (bioavailable and mobile) and as carbonate, oxide, and organic forms (fixed and immobile) (Peijnenburg et al., 2007). Accordingly, examining the correlations between soil physicochemical properties, especially the Fe fractions and Cd fractionation in soils, can be useful for elucidating the mechanisms of Cd mobility and bioavailability.

The Lianhuashan tungsten mine, which is located in Shantou City and within Guangdong Province, is one of the largest tungsten mines in China. The paddy fields downstream of the mining area have received substantial influxes of mine drainage. In this study, 73 pairs of soil and rice plant samples were collected from rice paddy fields in the Lianhuashan tungsten mining area. A frequently used sequential extraction procedure was used to fractionate the soil Cd (Tessier et al., 1979). Additionally, the Cd contents were determined in different parts of rice plants. To identify the main factors affecting Cd availability in rice, especially the role of Fe redox cycling on Cd availability, this study aimed to assess the effects of Fe fractions and other soil physicochemical properties, such as pH, OM, soil P, and CEC, on the distributions of soil Cd and on the availability of Cd for rice plants under field conditions.

2. Materials and methods

2.1. Sampling

Soil and plant samples were collected from 73 different rice paddy fields downstream of the Lianhuashan tungsten mine in November 2010 (Fig. S1). The rice plants were separated into grain (brown rice), husk, straw, and root fractions. Rice straws and roots were washed with tap water and then rinsed with deionized water more than three times. After being dried, all plant materials were ground in a carnelian mortar before further chemical analysis. The soil samples were collected from the root zone of the rice samples, air-dried, and then crushed with a wooden hammer before passing through a 2 mm sieve for pH analysis and a 100-mesh sieve for Cd and the other physicochemical analyses. Further descriptions and details regarding the sampling information are provided in a

previous study (Liu et al., 2014a).

2.2. Physicochemical analyses

To measure the soil pH, 1 g of air-dried soil was extracted with 1 M CaCl₂ (pH = 6.0) and then determined using a pH meter. The soil CEC was calculated as the sum of Ca + Mg + K + Na extracted with 1 M NH₄-acetate. The OM content was determined using the potassium dichromate volumetric method. The available phosphorus and total phosphorus contents were measured using the Mo–Sb anti-spectrophotography method. The contents of soil Fe fractions including DCB-extractable Fe, amorphous Fe, HCl-extractable Fe(II) and Fe(III) have been reported in a previous study (Liu et al., 2014a). The detailed soil physicochemical analysis procedures were described previously (Lu, 2000).

2.3. Cd Content in the soils and rice plants

Approximately 0.5 g of each soil sample was digested in 5 mL of HNO₃ and HClO₄ (4:1, v:v) at 140–160 °C. In addition, approximately 1 g of each plant sample was digested in 1.0 mL of HClO₄, 1.5 mL of H₂SO₄ and 4.0 mL of HNO₃ at 110–130 °C until a clear solution was obtained. All of the digested samples were adjusted to a constant volume depending on the content of Cd in the samples with 2% HCl, passed through a 0.45 μm filter and then analyzed for the total Cd using a graphite furnace atomic absorption spectrometer (ZEEnit 60, Analytik Jena, Germany). All reagents were of analytical grade or better and obtained from Guangzhou Chemical Co., China. To verify the accuracy of the metal analysis, certified reference materials, obtained from the National Research Center for Standards in China were used (GBW 07429 for the soils and GBW 10023 for the plants). The certified Cd contents in the GBW 07429 and GBW 10023 reference materials were 0.21 ± 0.02 and 0.57 ± 0.05 mg kg⁻¹, respectively. The results of our study (n = 12) provided mean Cd contents of 0.20 ± 0.03 and 0.61 ± 0.03 mg kg⁻¹ for the GBW 07429 and GBW 10023 materials, which corresponded to 95% and 107% of the target values, respectively. Reagent blanks and analytical duplicates were used to ensure the accuracy and precision of the analysis.

2.4. Soil Cd fractionation

A sequential Cd fractionation scheme based on a previous method (Tessier et al., 1979) was used to determine the following operationally defined Cd fractions: F1, Exchangeable fraction, adding 20 ml 1.0 M MgCl₂ (pH = 7.0) to 1 g soil and shaking for 4 h, centrifugal separation at 3000 r/min for 15 min, collecting the supernatant and then adding 10 ml 1.0 M MgCl₂ (pH = 7.0) to the precipitate and mixing on a rotary shaker, centrifugal separation, collecting the supernatant, repeating this procedure for three times and combining all the supernatants, adjusting to 50 mL with water and measure using a graphite furnace atomic absorption spectrometer; F2, Bound to carbonate fraction, adding 10 ml 1 M NaOAc/HOAc (pH = 5.0) to the precipitate from F1 and shaking for 6 h, centrifugal separation at 3000 r/min for 15 min, collecting the supernatant and adding 10 ml 1 M NaOAc/HOAc (pH = 5.0) to the precipitate and mixing on a rotary shaker, centrifugal separation, collecting the supernatant, repeating this procedure for three times and combining all the supernatants, adjusting to 50 mL with water and measure using a graphite furnace atomic absorption spectrometer; F3, Fe and Mn oxide bound fraction, adding 20 ml 0.1 M NH₂OH–HCl to the precipitate from F2 and shaking for 6 h, centrifugal separation at 3000 r/min for 15 min, collecting the supernatant and adding 10 ml 0.1 M NH₂OH–HCl to the precipitate and mixing on a rotary shaker, centrifugal separation, collecting the

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