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A three-season field study on the *in-situ* remediation of Cd-contaminated paddy soil using lime, two industrial by-products, and a low-Cd-accumulation rice cultivar

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ABSTRACT

To mitigate the serious problem of Cd-contaminated paddy soil, we investigated the remediation potential of combining *in-situ* immobilization with a low-Cd-accumulation rice cultivar. A three-season field experiment compared the soil pH, available Cd and absorption of Cd by three rice cultivars with different Cd accumulation abilities grown in Cd-contaminated paddy soil amended with lime (L), slag (S), and bagasse (B) alone or in combination. The three amendments applied alone and in combination significantly increased soil pH, reduced available Cd and absorption of Cd by rice with no effect on grain yield. Among these, the LS and LSB treatments reduced the brown rice Cd content by 38.3–69.1% and 58.3–70.9%, respectively, during the three seasons. Combined with planting of a low-Cd-accumulation rice cultivar (Xiang Zaoxian 32) resulted in a Cd content in brown rice that met the contaminant limit (≤0.2 mg kg^{-1}). However, the grain yield of the low-Cd-accumulation rice cultivar was approximately 30% lower than the other two rice cultivars. Applying LS or LSB as amendments combined with planting a low-Cd-accumulation rice cultivar is recommended for the remediation of Cd-contaminated paddy soil. The selection and breeding of low-Cd-accumulation rice cultivars with high grain production requires further research.

1. Introduction

Increasing anthropogenic activities, such as mining, industrial emissions and the application of sewage sludge and phosphate fertilizers, have caused severe heavy metal pollution in agricultural soils around the world (Wei et al., 2009; Zhu et al., 2012a; Lin et al., 2016). In China, approximately 19.4% (equivalent to approximately 2.0×10^7 ha) of agricultural soils are contaminated based on Chinese soil environmental quality limits (EQL), mainly with heavy metals (Ministry of Environmental Protection P. R.C. and Ministry of Land and Resources P. R.C. 2014). Among these contaminants, cadmium (Cd) ranks first in the percentage of soil samples (7.0%) exceeding the soil EQL. Soil-borne Cd is potentially one of the most harmful heavy metals to human health due to its facile transfer through the soil-crop system (di Toppi and Gabbrielli 1999; Chen et al., 2016). Notably, more than 10% of the brown rice in China was found to be contaminated with Cd (Li and Xu, 2015).

To mitigate the serious problem of Cd contamination, a number of options have been proposed to remediate Cd-contaminated soils, including immobilization, excavation and dumping, soil washing and flushing, and electrokinetics (Li and Xu, 2015). Among these methods, the immobilization of Cd in contaminated soils using inexpensive amendments is a fast and cost-effective *in-situ* remediation technique (Zhu et al., 2010; Feng et al., 2013). Lime and industrial by-products, such as slag, sugarcane bagasse and red mud, are widely used as amendments to immobilize Cd in contaminated soils. For example, lime applied at 150 g per m^2 significantly increased the soil pH and the form of carbonate and Fe/Mn oxides bounded Cd, decreased the Cd contents in plant tissues by 20–37.5% (Zhu et al., 2010). Slag added into soil at rates of 0.2–0.6% (w/w) significantly reduced the accumu-

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lation of Cd in the edible parts of crops by 12–90%, and the immobilization effect may have contributed to the increase in the pH and available Si in the soils (Chen et al., 2000). The addition of sugarcane bagasse can also immobilize Cd mainly through increases in the weak/unstable binding forces between the Cd and soil and in the strong forces of surface complexation (Tan et al., 2015). However, in some cases the Cd concentration in the edible parts of crops planted in Cd-contaminated soils may not meet the permissible agricultural product safety standard values after the application of amendments. Our previous study found that the application of lime and lime mixed with sepiolite to Cd-contaminated paddy soil significantly decreased the Cd contents in brown rice by more than 20%, but the Cd concentrations were still much higher than the quality standards (Zhu et al., 2010).

To further reduce the accumulation of Cd in brown rice, *in-situ* immobilization may be enhanced by other measures, such as planting low-Cd-accumulation cultivars of plants. Previous studies have indicated a wide variation in the Cd uptake and accumulation of different rice cultivars (Chen et al., 2016). A field experiment conducted in a low-Cd paddy soil indicated that there was a 14-fold difference in the grain Cd concentration among 110 tested hybrid rice cultivars, and super rice cultivars exhibited significantly higher grain Cd uptake than the common hybrid rice cultivars (Shi et al., 2009). Similar differences were also observed in the Cd accumulation ability of 158 newly developed rice cultivars grown in twelve locations, with higher grain Cd concentrations detected in hybrid rice cultivars compared to conventional rice cultivars (Cao et al., 2014). Moreover, based on the regression models developed for the average *Indica* and *Japonica* rice cultivars, it is clear that *Indica* rice is more likely to accumulate Cd in its grains than *Japonica* rice (Römkens et al., 2009; Zhao et al., 2015). However, there is limited information on the effect of combining amendments with low-Cd-accumulation cultivars on the Cd accumulation in rice grains.

Therefore, a two-year field experiment encompassing three rice seasons was conducted to investigate the effects of three amendments (lime, slag and bagasse) and mixtures thereof combined with three rice cultivars on the accumulation of Cd in rice.

2. Materials and methods

2.1. Site description

The field trial was conducted in Suxian County, Hunan Province (N, 25° 30′21″; E, 112° 53′55″) based on the investigation of both soil and brown rice that taken in the first half year of 2013. Thus, the three seasons experiment was conducted with the funding of 2-year project. The climate of the study area is warm, humid, and subtropical with a mean annual temperature of 18.0 °C and an average total annual precipitation of 1490 mm. The tested soil was derived from Quaternary red clay, is classified as acidic Ultisol, and has been used to cultivate double rice for many years. During the 1970s and 1980s, a large amount of heavy-metal-rich lignite was used as an organic fertilizer, which caused widespread pollution of the local rice production fields, with Cd as the primary heavy metal contaminant. The soil basic properties were as follows: pH, 5.72; organic carbon, 34.2 g kg^{-1}; total nitrogen, 2.68 g kg^{-1}; available phosphorus, 26.9 mg kg^{-1}; cation exchange capacity (CEC), 15.7 cmol kg^{-1}; clay, 48.9%; total Cd, 1.2 mg kg^{-1} and $CaCl_2$-extractable Cd, 0.053 mg kg^{-1}.

2.2. Experimental design and treatments

There were a total of 8 treatments, denoted as CK, no amendment applied; L, lime applied; S, slag applied; B, bagasse applied; LS, mixture of lime and slag applied; LB, mixture of lime and bagasse applied; SB, mixture of slag and bagasse applied; LSB, mixture of lime, slag and bagasse applied. Each of the treatments had three replicates.

Table 1
The contents of heavy metals and pH of the amendments.

Amendments	pH	Heavy metals (mg kg^{-1})					
		Cd	Cu	Cr	Pb	Zn	Ni
Lime	12.75	0.24	1.17	1.00	0.98	5.82	1.62
Bagasse	10.14	0.76	21.87	2.81	4.42	47.68	1.06
Slag	9.08	1.20	76.12	4.19	2.03	322.51	9.19

The plots were 8 m×5 m in area and these were randomly arranged in three blocks, each containing a replicate of the treatments. The blocks were separated by 0.8-m ditches and the adjacent plots were separated with a ridge (0.3 m high and 0.3 m wide) covered with plastic film. Before the 24 plots were separated, the field was tilled to a depth of 15–20 cm. Due to our previous study, the lime (purchased from the Taoyuan County Ligonggang Lime Corp.), slag (Longping High-Tech Fertilizer Co. Ltd.) and bagasse (Guangxi Liuzhou City Sugar-refinery Co. Ltd.) were applied once at 90, 1125 and 1125 g m^{-2} (Huang et al., 2000). The pH and heavy metal contents (Cd, copper (Cu), chromium (Cr), lead (Pb), zinc (Zn) and nickel (Ni)) of the amendments are provided in Table 1. The amendments were hand-scattered over the surface soil and mixed thoroughly by manual ploughing to a depth of 10–15 cm before the transplanting of rice seedlings for the first season.

For the first season (2013 late rice season), a hybrid *indica* cultivar (local commonly cultivar) named Y Liangyou7 (YL7) was planted. Due to the high Cd accumulation potential of YL7 and the different growth periods of early rice and late rice, two conventional *indica* cultivars named Xiang Zaoxian32 (XZ32) and Xiang Wanxian12 (XW12) were used as the early and late rice cultivars, respectively, in 2014. Seven days after the amendments were applied (the first season) and two days after the base fertilizers applied (the latter two seasons), 30-d-old rice seedlings were transplanted at a spacing of 0.20×0.25 m with three seedlings per hill for early rice and one seedling per hill for late rice. Transplanting was carried out manually in each season. All the field management practices followed the local practice. The specific compound fertilizer for rice was used as a base fertilizer (N:P:K =16:8:16), applied to the soil at 900 kg ha^{-1} two days before transplanting, and urea was used as a topdressing fertilizer, applied to the soil at 30 kg N ha^{-1} seven days after transplanting. The field water regime was managed as an alternating cycle of flooding, drainage and intermittent wetting during the stages of seedling, tillering, and spiking/ripening, respectively, throughout the whole rice growing season.

2.3. Sampling

Soil samples were collected from each plot before the application of amendments (for the analysis of the basic properties) and after the harvest in each season. Each soil sample was a composite of approximately 8 cores collected randomly from the experimental plot at a depth of 0–20 cm. Air-dried samples were sieved through a 2 mm screen, homogenised and preserved. Rice was harvested at the maturity stage (border rows removed and the remainder harvested), at which time the fresh weights of grain and straw in each plot were recorded. Rice grain and straw samples were composites of approximately 8 sample points collected randomly from the experimental plot. Then, the water contents of the rice grain and straw samples were measured by oven drying at 60 °C for 48 h. The rice grain yield of each plot was calculated from the fresh grain weight and water content. After oven drying, the rice grains were separated into brown rice and rice hulls. Finally, the brown rice, rice hull and rice straw samples were pulverised and preserved.

2.4. Analysis

Aqua regia-$HClO_4$ was used for digestion to determine the total