THE REDUCTION OF WHEAT Cd UPTAKE IN CONTAMINATED SOIL VIA BIOCHAR AMENDMENT: A TWO-YEAR FIELD EXPERIMENT

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A field study involving wheat production was extended in order to study the effects of biochar (BC) amendment in paddy soil that had long-term contamination of Cd. The BC was used as an amendment in Cd-contaminated soil for its special property. BC was amended at rates of 10 to 40 t ha^{-1} during the rice season before rice transplantation in 2009. BC amendments increased soil pH by 0.11 to 0.24 and by 0.09 to 0.24 units, respectively, while the soil $CaCl_2$-extracted Cd was reduced by 10.1% to 40.2% and by 10.0% to 57.0% in 2010 and 2011, respectively. Consequently, the total wheat Cd uptake was decreased by 16.8% to 37.3% and by 6.5% to 28.3%. Wheat grain Cd concentration was reduced by 24.8% to 44.2% and by 14.0% to 39.2% in 2010 and 2011, respectively. The BC application in soil reduced Cd phyto-availability in two wheat seasons possibly by raising soil pH and soil organic carbon (SOC). Therefore, BC may be used for soil remediation, but not to reduce Cd uptake to an adequate level for food production on Cd contaminated soils.

Key words: Biochar (BC); Cd; Wheat; Heavy metal contamination; Soil amendment

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INTRODUCTION

Cadmium (Cd) pollution in croplands has been a serious concern for subsistence farmers due to the risk of soil-food chain transfer (Chaney *et al.* 2005). Potential health problems caused by chronic Cd exposures are numerous and frequently reported (Rignell-Hydbom *et al*. 2009). Wheat is considered to be one of the major crops having high potential to accumulate Cd (Bose and Bhattacharyya 2008). Even small increases in the Cd content of grain could have long-lasting and widespread harmful impacts on the well-being of consumers (Singh *et al*. 2010). Worldwide, the areas containing Cd pollution have been expanding, and a number of soil pollution studies have also found that increasing areas of cropland in China contain Cd pollution (Pan *et al*. 1999). Vast areas of rice paddies in the Yangtze River valley of China have also been reported to be contaminated with Cd (Zhang *et al*. 2009), and Cd content appears in an increasing trend in rice grains randomly collected from paddy soil in China (Zhen *et al*. 2008). This has raised concerns for farmers living on rice and wheat. Therefore, in order to ensure food safety for consumers, measures to mitigate the Cd pollution in croplands and to decrease Cd levels in crop grains are urgently needed.

There have been many studies on alleviating Cd mobility and thus reducing Cd uptake by crops using soil amendments such as alkaline materials, limestone, and organic matter (Madejón *et al*. 2009; Baker *et al*. 2011; Garau *et al*. 2007). These amendments have been observed to manipulate soil pH and/or increase the soil organic carbon (SOC) content, which stabilizes the readily mobile Cd levels of the soil (Bradl 2004). In particular, the study by Garau *et al*. (2007) found that the soil pH could be raised from 4.23 to 7.11, and that $Ca(NO_3)_2$-extracted Cd decreased by 98.6% after addition of 40 g red mud per kg soil in a loamy-sand soil. In the study by Madejón *et al*. (2009), the soil $CaCl_2$-extractable Cd was decreased by 46.5%, while soil pH (H_2O) and SOC increased by 2 units and 3.5 g kg^{-1}, respectively, after four years of continuous amendment of compost biosolid (60 g kg^{-1}) to a Typic Xerofluvent soil. While these amendments could be considered a more efficient way to decrease Cd mobility in soils (Kumpiene *et al*. 2008), commercial amendments are expensive. Therefore, the development of cost-effective soil amendments is a priority in soil environmental technology for ensuring Cd safety while sustaining crop production in the croplands.

For the last decade, the agricultural application of BC produced via crop biomass pyrolysis has been strongly recommended as an approach to enhance soil C stock and N retention, as well as to improve soil fertility (Sohi *et al.* 2010; Major *et al.* 2010; Lehmann *et al*. 2011). BC can be produced via oxygen-limited pyrolysis at temperatures varying between 300 and 1000 °C from various crop residue feedstocks (Meyer *et al.* 2011). BC contains a large amount of recalcitrant organic material, and the major carbon component is composed of aromatic groups (Shih *et al.* 2012; Harvey *et al.* 2012). Recent studies have also reported that BC is an effective soil amendment for heavy metal-contaminated soil (Beesley *et al.* 2011a; Cui *et al*. 2011; Cao and Harris 2010). BC was found to enhance Cu and Pb sorption in soil (Trakal *et al*. 2011). Park *et al*. (2011) also showed in an incubation experiment that BC application was effective at the immobilization of Cd, Pb, and Cu. The BC was also found effective for the immobilization of heavy metals in short-term incubation experiments (Jiang *et al.* 2012; Borchard *et al*. 2011; Bradl 2004). The crop biomass used as a feedstock for BC is taken from agriculture waste. For this reason, BC is anticipated to be a low-cost and multiple-benefit amendment for economical contaminated soil treatment.

In the previous study, considerable effects of the BC amendment on the immobilization of soluble Cd and on the reduction of rice Cd uptake in a long-polluted flood field were observed (Cui *et al*. 2011). However, it is still not known whether the BC amendment is also applicable to wheat crops under un-irrigated conditions. The present study looks into the effects of BC on soil Cd mobility and on wheat uptake in a field experiment lasting for two consecutive years in a long-term polluted paddy field. The possibility of effectively using a BC amendment to alleviate Cd pollution and to reduce the Cd health risk associated with wheat in polluted croplands for purposes of supporting China's sustainable agriculture is addressed.

EXPERIMENTAL

Experiment Design

The experiment was conducted in Yixing (31º24'26"N, 119º41'36"E), Jiangsu, China where heavy metals emitted by a smelter had contaminated the soils in the 1970's.

The paddy soil is ferric-accumulic stagnic anthrosols (Gong 1999) and is under long term summer rice (*Oryza sativa* L.) – winter wheat (*Triticum aestivum*) rotation. BC was a fine powder produced via pyrolysis of wheat straw at 350 °C to 550 °C using a vertical kiln made of refractory bricks at the Sanli New Energy Company, Henan Province, which was an uncontaminated location. The BC properties were pH(H_2O) 10.35, total carbon content 467.2 g kg^{-1}, CEC 18.05 cmol kg^{-1}, and total Cd 0.03 mg kg^{-1}, respectively (Cui *et al.* 2011). The produced BC was ground through a 2-mm sieve. The field experiment was initiated in 2009. There were four BC application rates at 0, 10, 20, and 40 t ha^{-1}, respectively. BC was amended before rice transplantation only in this year. BC was added into the soil by plowing and raking to a depth of 0 to 15 cm. As a conventional farming system of the region, winter wheat was sowed after rice harvest in late October in each year. For wheat production, the seeds of the cultivars Ningmai-13 and Zhenmai-5 were sowed in 2009 and in 2010, respectively. Before each sowing, the basal fertilizers N, P_2O_5, and K_2O were applied consistently both years of the experiment at rates of 120, 125, and 125 kg ha^{-1}, respectively, in the forms of urea, $Ca(H_2PO_4)_2$, and KCl, respectively. The crop production management was kept consistent through the experimental plots. The experimental plots were defined as areas of 4 m × 5 m and were arranged in a randomized complete block design with three replicates.

Soil and Wheat Sampling Collecting

Topsoil (0 to 15 cm) sampling was done before rice sowing and after the wheat harvest in May of both 2010 and 2011. Three *S*-shaped undisturbed core samples were collected from each plot using a soil core sampler (Eijkelkamp, Netherlands). All the soil samples were air-dried, cleared of plant detritus and any visible fragments, and ground to pass through a 2-mm sieve for soil pH, $CaCl_2$, and DTPA Cd extraction. A portion of each sample was further ground to pass through a sieve of 0.15 mm for SOC, CEC, and total Cd. The soil basic properties pH(H_2O), total carbon, CEC, and total Cd were 5.60, 20.71 g kg^{-1}, 18.05 cmol kg^{-1}, and 21.84 mg kg^{-1}, respectively (Cui *et al.* 2011).

In brief, soil pH (H_2O) was measured using in a soil-to-solution ratio of 1:2.5. Total Cd contents were determined by digesting the 0.5000 g soil with a mixed solution of HF, HNO_3, and $HClO_4$ (Lu 2000). Soil available Cd was estimated by extraction with 0.01 mol L^{-1} $CaCl_2$ and 0.005mol L^{-1} DTPA (Lu 2000).

At harvest, dry wheat biomass and grain yield for each plot were measured. Before this, three composite plant samples were randomly collected from a single plot on the 20th of May of both years, which was during the ripening stage. The samples were washed in order to remove attached soil particles, first with tap water and then with deionized water. Each plant sample was then separated into roots, shoots, and grains, which were then all dehydrated in an air-convection oven at 105 °C for 30 min and further dried to constant weight at 60 °C for another 48 hours (Lu 2000). The dried samples were crushed, mixed, homogenized, and stored in air-tight polyethylene bags prior to chemical analysis.

Data Processing and Statistics

All data were expressed as means plus or minus one standard deviation. Differences between treatments were examined by two-way analysis of variance (ANOVA). All statistical analyses such as regressions of linear and nonlinear significant difference ($p<0.05$) were carried out using SPSS, version 13.0 (SPSS Institute, USA, 2001).

RESULTS

Soil Cd Mobility

Compared to the unamended soil, the pH of the soil with the BC amendment was significantly higher, at a mean rate of increase by 0.01 units per ton of BC amended consistently over the two years (Table 1). The soil pH was significantly lower than that in the rice season reported in a previous work (Cui *et al*. 2011). Likewise, the SOC contents were higher than those of unamended soil by 10.0%, 48.0%, and 57.0% in 2010 and by 9.4%, 22.5%, and 42.3% in 2011 for the BC amendment rates of 10, 20, and 40 t ha^{-1}, respectively (Table 1).

Table 1. Changes in Soil pH, SOC, and Concentrations of $CaCl_2$- and DTPA-Extracted Cd from Soil with BC Amendment

BC application rates (t ha^{-1})	Soil pH (H_2O)	SOC (g kg^{-1})	$CaCl_2$ extracted Cd (mg kg^{-1})	DTPA extracted Cd (mg kg^{-1})
	2010			
0	5.51±0.12b	21.55±0.36b	6.01±0.34a	14.77±0.82a
10	5.61±0.06ab	23.70±3.04b	5.40±0.36ab	14.29±0.42a
20	5.69±0.16ab	31.89±0.17a	4.60±0.30b	13.00±0.55b
40	5.76±0.09a	33.83±2.51a	3.60±0.71c	13.13±0.43b
	2011			
0	5.58±0.06b	22.67±0.22c	3.56±0.48a	16.98±2.07a
10	5.70±0.08ab	24.81±1.45c	3.50±0.53a	14.32±1.38ab
20	5.81±0.09a	27.76±1.16b	2.74±0.69a	14.90±1.59ab
40	5.86±0.08a	32.26±2.10a	2.77±0.24a	12.50±0.07b

Different lower case letters in a single column represent significant differences between the treatments in a single year ($p < 0.05$; $n = 3$; mean ± S.D.).

As shown in Table 1, $CaCl_2$-extracted Cd was 10.1%, 23.4%, and 40.2% lower than the control in 2010 and 1.8%, 23.1%, and 22.1% lower than the control in 2011 for the 10, 20, and 40 t ha^{-1} treatments, respectively (Table 1). As a large pool of the total Cd, the DTPA-extracted Cd concentration from the BC-amended soil followed a trend similar to that of the $CaCl_2$-extractable Cd. However, no reduction in DTPA-extractable Cd that was proportional to the BC amendment rate was observed.

Cd Uptake and Partitioning by Wheat

The data in Table 2 shows that grain yield and biomass of wheat were not significantly different between the treatments of a single year, but they were higher in 2011 than in 2010 under BC application. The total Cd uptake of the wheat plant was significantly reduced by the consistent BC amendment over the two year period (Fig. 1, $p = 0.0399$ for 2010; $p = 0.0012$ for 2011). Total Cd uptake (mg ha^{-1}) was calculated for both years and showed the effect of BC on reducing Cd uptake in this area (Fig. 2). Compared to the control, the total plant Cd uptake was 16.8%, 27.1%, and 37.3% lower in 2010 and 6.5%, 19.4%, and 28.3% lower in 2011 under the 10, 20, and 40 t ha^{-1} BC application rates, respectively. A large portion, 30 to 40%, of the total Cd uptake was seen to decline in 2010 and in 2011 under 40 t ha^{-1} of BC amendment.

Table 2. Wheat Grain Yields and Biomass with BC Amendment

BC Application rates (t ha^{-1})	Grain yield (t ha^{-1})		Biomass of wheat (t ha^{-1})	
	2010	2011	2010	2011
0	2.45±0.47a	5.31±0.04a	6.39±1.23a	13.87±0.10a
10	2.00±0.13a	5.62±0.39a	6.36±0.41a	14.66±1.02a
20	2.49±0.71a	5.66±0.56a	6.51±1.86a	14.79±1.47a
40	2.53±0.39a	5.91±0.37a	6.61±1.02a	15.43±0.96a

Different lower case letters in a column indicate significant differences between the treatments in a single year ($p < 0.05$; n = 3; mean ± S.D.).

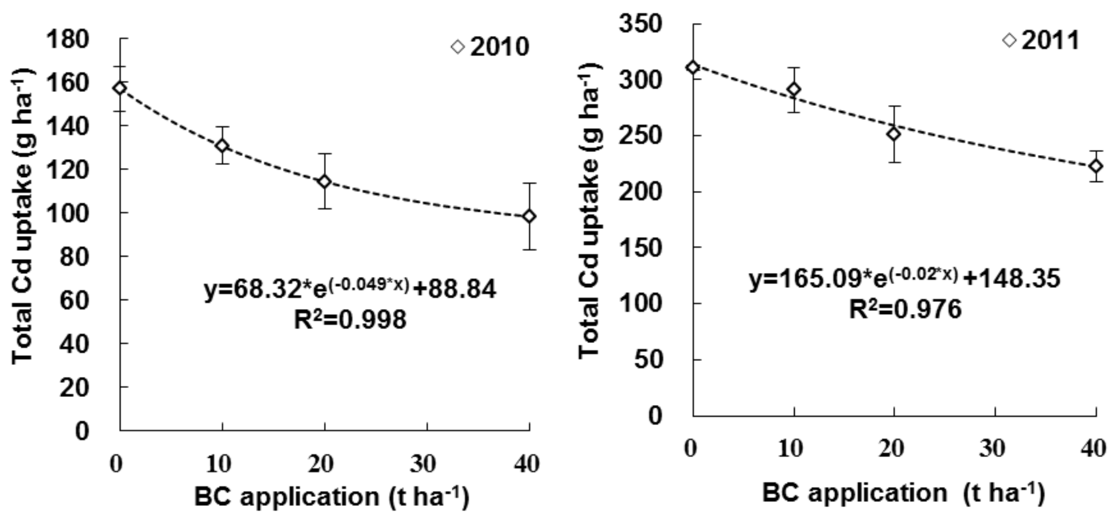

Fig. 1. Total Cd uptake by wheat plant in relation to BC amendment rates

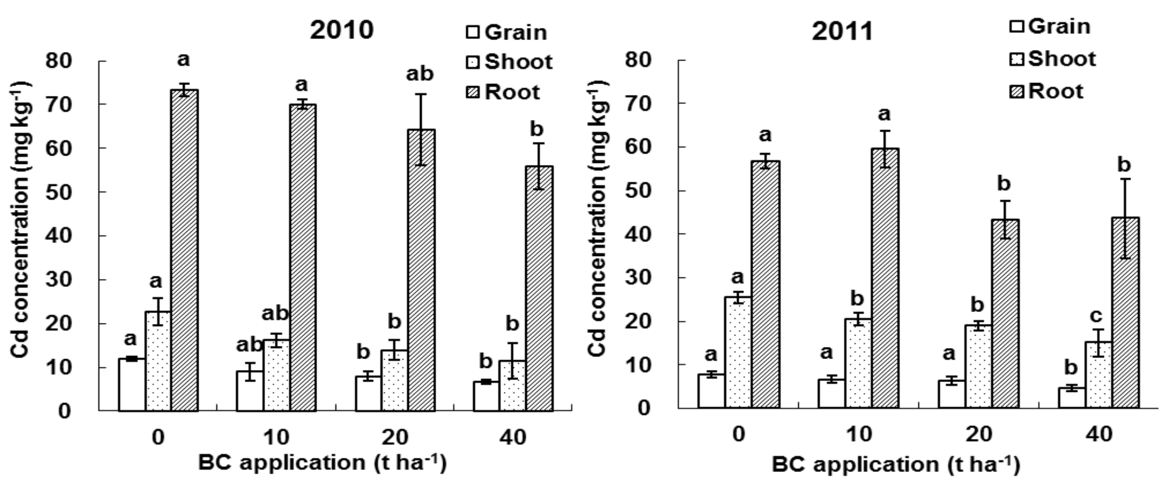

Fig. 2. Cd concentrations in plant tissues in BC amended soil (the different lower case letters indicate a significant difference between the treatments in a single year, (p < 0.05; n = 3; mean ± S.D.)

Figure 2 shows the Cd concentrations of the plant tissues under BC amendment treatments. Compared to the control, the wheat grain Cd concentrations were reduced by 24.8%, 33.3%, and 44.2% in 2010 and by 14.0%, 17.4%, and 39.2% in 2011 under the BC amendments of 10, 20, and 40 t ha^{-1}, respectively. On average, the Cd concentrations of the wheat grains, shoots, and roots were reduced by 0.57, 1.21, and 0.30 mg kg^{-1}, respectively, in 2010 and by 0.32, 2.02, and 0.61 mg kg^{-1}, respectively, in 2011 per ton of BC amended.

DISCUSSION

Influence on Soil Properties and Available Cd

The results showed that BC treatments at rates between 10 and 40 t ha^{-1} significantly decreased the concentrations of soil Cd extracted by $CaCl_2$ and DTPA, which was similar to the results for rice season soil (Cui *et al*. 2011). The amount of BC application accounted for about 0.4% to 1.7% of the surface soil in our experiment, and the total soil Cd concentration decreased by 1.2% to 5.7% with BC application. However, it was found that, with the exception of the 10 t ha^{-1} treatment, with BC application, the soil $CaCl_2$-extracted Cd concentration decreased by more than 10%, and the DTPA-extracted Cd decreased by 11%. This indicated that there was a physiochemical reaction involving the inhibition of Cd phytoavailability by BC. In general, the amount of Cd^{2+} sorbed on the surface of soil colloids increases as the soil pH increases (Bradl 2004). Whereas the concentrations of both $CaCl_2$- (2010, $p = 0.02$; 2011, $p = 0.08$) and DTPA-extracted (2010, $p = 0.07$; 2011, $p = 0.13$) Cd were negatively correlated to pH both years (Figs. 3 A and B), the total Cd uptake by wheat plant was found to be in a linear correlation with both SOC content (2010, $p = 0.05$; 2011, $p = 0.002$) and soil pH (2010, $p = 0.01$; 2011, $p = 0.03$) for both years (Figs. 3 C and D). Again, the Cd concentration of wheat grain was linearly correlated with the $CaCl_2$- (2010, $p = 0.06$; 2011, $p = 0.24$) and DTPA-extracted (2010, $p = 0.11$; 2011, $p = 0.04$) Cd contents taken from the amended soil (Figs. 3 E and F). However, a positive relationship of grain Cd and $CaCl_2$ extractable Cd under BC treatments was seen in both years (Fig. 3 E).

It was already known that the $CaCl_2$ extractability provides a sound indication of phytoavailability for metals under evaluation of Cd plant uptake (Cui *et al*. 2011; Van Erp *et al*. 1998; Zhu *et al*. 2012). For the contaminated soils, the DTPA-extracted Cd was higher than the $CaCl_2$-extracted soil both years, as the two extracting reagents have different chemical matrices and will thus extract different forms of Cd from the soils. The SOC increase under the BC amendment also played a major role in decreasing the plant Cd uptake (Fig. 3 E). This is possibly because the BC increased the SOC pool (Beesley *et al.* 2011b).

Wheat Yield and Cd Uptake

There were no significant differences in this experiment with regard to the yields of wheat grain and rice grain in the soils with BC treatments in 2009 and 2010. Therefore, it is concluded that BC improved soil properties but had no effect on plant growth. Increased crop yield is a commonly reported benefit of adding BC to soils, though experimental results are variable and dependent on the experimental set-up, soil properties, and conditions (Jeffery *et al.* 2011).

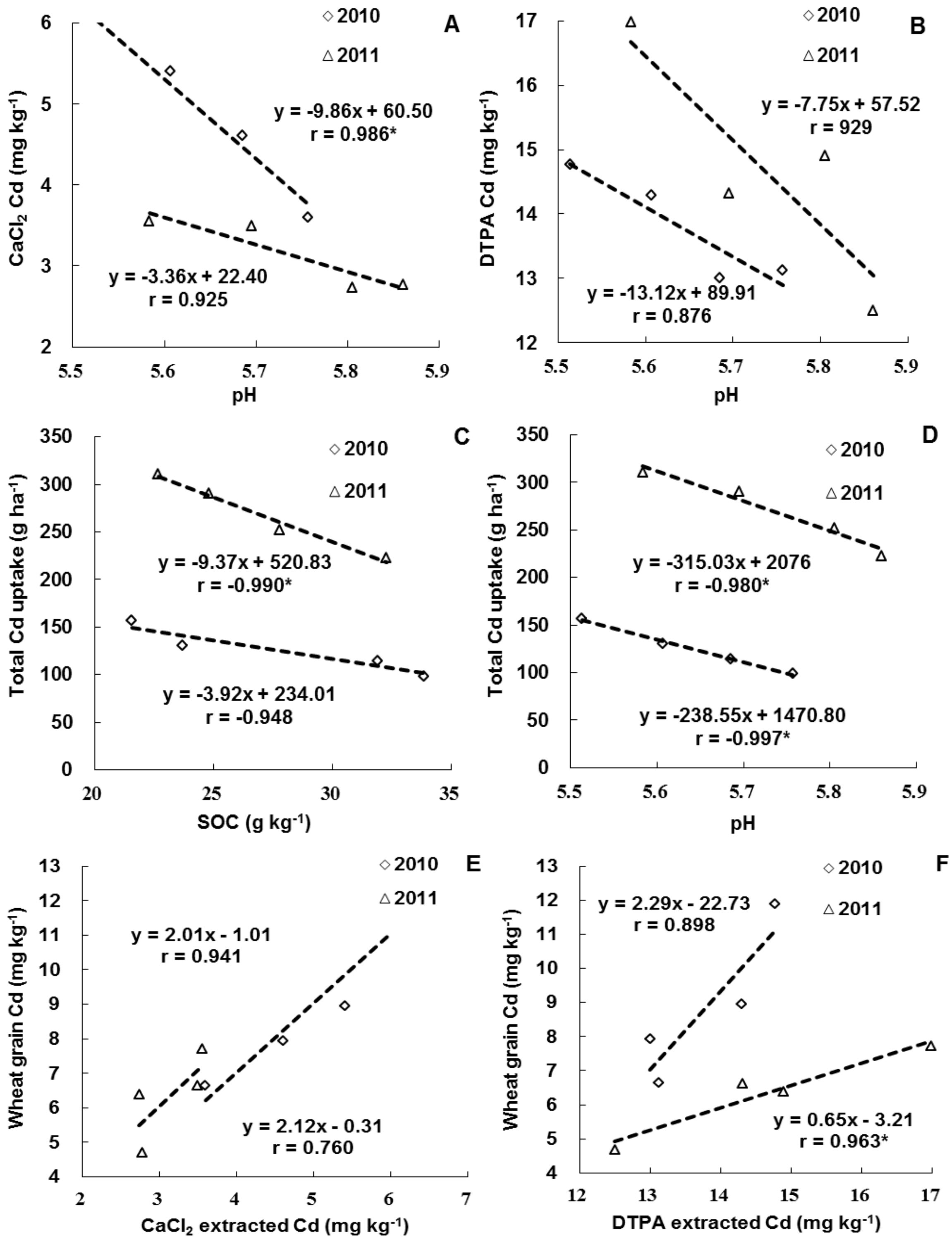

Fig. 3. Correlations: $CaCl_2$- (A) and DTPA- (B) extracted soils Cd vs. pH; total Cd uptake vs. SOC (C) and pH (D); and $CaCl_2$- (E) and DTPA- (F) extracted soils Cd vs. Cd concentration of wheat grain

Glaser *et al*. (2002) reported that 0.5 t ha^{-1} BC increased the yield of cowpeas, and more a recent investigation in Italy conducted with durum wheat (*Triticum durum* L.) found that the yield was increased by 10% after the addition of 10 t ha^{-1} of BC (Baronti *et al*. 2010). Vaccari *et al*. (2011) reported that BC had no negative consequences on crop yield when the BC, which had a pH (H_2O) 7.2 and was amended at rates of 30 t ha^{-1} and 60 t ha^{-1}, was used in a field experiment in 2009 and 2010.

According to the results, the concentrations of wheat grain Cd were reduced by 24.8%, 33.3%, and 44.2% in 2010 and by 14.0%, 17.4%, and 39.2% in 2011 under the BC amendment of 10, 20, and 40 t ha^{-1}, respectively, although the Cd concentrations in the wheat still far exceeded the limit set by China National Standards (0.2 mg kg $^{-1}$). Similar trends of Cd content in rice grain were observed in this field (Cui *et al*. 2011). The variation of Cd uptake with cultivars had been well demonstrated in previous studies (Li *et al*. 2005). In this study, the wheat grain Cd of the cultivar Ningmai-13, a comparatively low Cd uptake cultivar, was found to be reduced as low as 6.64 mg kg^{-1} under the BC amendment at 40 t ha^{-1}, in comparison to 11.89 mg kg^{-1} without BC. Thus, the BC amendment, in combination with low Cd cultivars breeding, could offer a basic means of producing low-Cd wheat in contaminated soils, a goal that has been emphasized for China's cereal production, especially with regards to rice agriculture, in order to decrease the health risks associated with Cd food exposure.

In the results of this study, the wheat grain Cd concentration was 3.41 to 7.38 times higher than the rice grain, and the $CaCl_2$- and DTPA-extracted Cd concentrations in the wheat season were 2.34 to 4.39 times and 1.05 to 1.62 times greater, respectively, compared to those observed in the rice season in the BC treatment field experiment of Cui *et al*. (2011). Besides the different species, the different water management could have affected the Cd forms in the contaminated soil. Continuous flooding in paddy fields has been found to significantly reduce the Cd content in brown rice (Jung and Thornton 1997) due to the involvement of hydroxide, carbonates, sulphide, and iron compounds under reducing conditions, while the reduced species were oxidized during the wheat season (de Livera *et al*. 2011). This resulted in significant increases in $CaCl_2$-extracted Cd in the wheat season compared to the rice season.

CONCLUSIONS

Over a two-year period, biochar (BC) effectively immobilized Cd in paddy soil and reduced Cd uptake by wheat in long-term contaminated dry paddy soils, and this reducing effect lasted two years. The Cd concentration of plant tissue was significantly reduced, especially in the grain, though this uptake did not reduce grain Cd contents to limits recommended for food production. Therefore, the BC amendment can be used as an ecological engineering technology for Cd-contaminated soil to diminish the risk of environmental pollution.

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