# RESEARCH ARTICLE

# Speciation and phytoavailability of cadmium in soil treated with cadmium-contaminated rice straw

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Received: 11 May 2014 /Accepted: 24 August 2014 /Published online: 9 September 2014  $\oslash$  Springer-Verlag Berlin Heidelberg 2014

Abstract When grown on Cd-contaminated soil, rice typically accumulates considerable Cd in straw, and which may return to the soil after harvest. This work was undertaken to assess the pollution risk of Cd associated to the Cdcontaminated rice straw after incorporating into an uncontaminated soil. With the Cd-contaminated rice straw added at 0, 1, 2, 3, 4 and 5 %  $(w/w)$ , an incubation experiment (28 days) with non-planting and a followed pot experiment sequent with two planting (rice and Chinese cabbage, transplanted after 28-day incubation) were carried out to investigate the changes of soil Cd speciation and phytoavailability. The results indicated that the Cd-contaminated rice straw addition significantly increased soil pH and dissolved organic carbon during the 28 day incubation. For the high availability of Cd in contaminated rice straw, diethylenetriaminepentaacetic acid (DTPA) extractable Cd significantly increased, and the percentages of acetic acid extractable and reducible Cd in soil significantly enhanced after the addition of Cd-contaminated rice straw. However, the Cd-contaminated rice straw addition inhibited the rice growth and induced the decrease of Cd in rice grain and straw by 12.8 to 70.2 % and 39.3 to 57.3 %, respectively, whereas the Cd contents increased by 13.9 to 84.1 % in Chinese cabbage that planted after rice harvest. In conclusion,

Responsible editor: Stuart Simpson

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Z.<C. Luo Hunan Soil and Fertilizer Institute, Changsha, Hunan, China Cd associated with Cd-contaminated rice straw was highly available after incorporating into the soil, and thus the Cd pollution risk via the Cd-contaminated rice straw incorporation should be evaluated in the Cd-contaminated paddy region.

Keywords Rice straw . Cadmium . Speciation . DTPA extractable Cd . Chinese cabbage

# Introduction

Cadmium is a toxic heavy metal of great environmental concern with unknown essential function for life. Several human activities, such as mining, smelting, disposing of sludge, fertilising, etc., may cause Cd contamination in farmland. There is approximately  $2.0 \times 10^7$  ha farmland (occupied 19.4 % of the total farmland) in China that has been contaminated by heavy metals, mainly Cd, which mostly distributes in Chinese paddy region (Ministry of Environmental Protection P.R.C. and Ministry of Land and Resources P.R.C. [2014\)](#page-6-0). The maximum Cd content in contaminated paddy field varies from 5.0 to 145.0 mg  $kg^{-1}$ , resulting in the maximum accumulation of Cd in brown rice at levels from 1.9 to 9.4 mg kg−<sup>1</sup> (Zhang [2008;](#page-7-0) Zhu et al. [2012a](#page-7-0)). The remediation of these contaminated paddy soil is implemented using various measurements and will last for years. During the process, these paddy soils remain in crop production, and the products are attempted to use as industrial raw materials. Accordingly, plenty of Cd-contaminated rice straw should be produced for rice straw that commonly accumulated considerable Cd. However, a large amount of Cd-contaminated rice straw is still returned to on-site paddy fields or abandoned and dispersed to adjacent environment in such region. For example, approximately 30 and 25 % of the annual produced crop straw (mainly rice straw) are utilised and abandoned, respectively, in the subtropical region of China (Zhu et al. [2005\)](#page-7-0). Therefore, the Cd risk related to Cd-contaminated rice straw incorporated into soil deserved concern and investigation.

Effects of crop straw incorporation on the speciation and phytoavailability of Cd in Cd-contaminated soils can be divided into two contradictory views. A part of the studies indicated that the addition of crop straw may reduce the availability of Cd via increase in the soil pH. For example, Cui et al. [\(2008\)](#page-6-0) conducted a 6-month incubation experiment and found that added 6 % rice straw (Cd content was not determined) to artificial Cd-contaminated acid soils significantly decreased the total soluble Cd concentration from 20 to 15 nmol  $L^{-1}$  and decreased the free Cd<sup>2+</sup> concentration from 16 to 12 nmol L−<sup>1</sup> . Li et al. [\(2014\)](#page-6-0) observed that application of rice straw (total Cd 0.08 mg  $kg^{-1}$ ) into mining contaminated soil at rates of 7.8 and 11.8 g  $kg^{-1}$  that significantly decreased acid extractable and reducible fractions of Cd in the soil and Cd concentrations in lettuce. However, the other part of studies reported that crop straw addition induced increase of soil dissolved organic carbon (DOC), which can contribute to enhancement of soil Cd availability. For instance, Wu et al. [\(2012\)](#page-6-0) found that rice straw (total Cd 0.49 mg  $kg^{-1}$ ) added into Cu smelter-contaminated soil at  $1\%$  (w/w) did not enhance the amount of water or NH<sub>4</sub>OAc extractable Cd in soil but significantly increased Cd concentrations in plant shoot from 197 to 365 mg  $kg^{-1}$  as compared to the control. Bai et al. [\(2013\)](#page-6-0) found that an additional 0.25 to 1.0 % wheat straw (Cd content was not determined) increased soluble Cd in artificial Cd-contaminated soil by 10 to 33 % and increased the Cd concentration in rice tissues by 120 %. As aforementioned, the incorporation of rice straw may induce the changes in soil pH and DOC content and thus affect the availability of Cd in soil. However, impacts of Cd in rice straw per se on Cd availability in soil are paid limited attention, although such information may be useful for management of Cd-contaminated rice straw. The ultimate objective of this study was to evaluate the pollution risk of Cd on soil and agricultural products by investigating the changes in the speciation and phytoavailability of Cd in an uncontaminated soil with the incorporation of Cd-contaminated rice straw.

#### Materials and methods

#### Soil and rice straw

Surface soil (0–20 cm) was collected from Taoyuan County, Hunan Province. The soil was air-dried and passed through a 10-mm mesh sieve for the incubation and pot experiment, and then a representative subsample was collected and passed through a 2-mm mesh sieve for a soil property analysis. The tested soil was classified as acidic Ultisols derived from Quaternary red clay. The soil characteristics were as follows: pH, 4.72; organic C, 13.13 g kg<sup>-1</sup>; total N, 1.33 g kg<sup>-1</sup>; total P, 0.97 g kg<sup>-1</sup>; total Cd, 0.37 mg kg<sup>-1</sup>; and clay, 36.4 %.

The Cd-contaminated rice straw was collected from Zhuzhou City, Hunan Province. The paddy field was heavily contaminated with Cd via the drainage of wastewater from a nearby electroplating factory (Zhu et al. [2010](#page-7-0)). The rice straw was oven-dried at 60 °C and passed through a 1-mm mesh sieve for the experiment and basic property analysis. The contaminated rice straw characteristics were as follows: organic C, 394.13 g  $kg^{-1}$ ; total N, 10.19 g  $kg^{-1}$ ; total P, 1.53 g kg<sup>-1</sup>; total Cd, 6.66 mg kg<sup>-1</sup>; water extractable Cd, 0.34 mg kg−<sup>1</sup> ; and diethylenetriaminepentaacetic acid (DTPA) extractable Cd, 6.35 mg  $kg^{-1}$ .

# Incubation and pot experiments

The Cd-contaminated rice straw was homogeneously mixed with soil at rates  $(w/w)$  of 0 (control, R0), 1 (R1), 2 (R2), 3  $(R3)$ , 4  $(R4)$  and 5 %  $(R5)$ . Three replicates of each treatment were tested. Each treated soil (7.0 kg) was then placed in plastic pots, 25 cm in height and 31 cm in diameter, and incubated outside for 28 days (2 June to 29 June 2011) in Changsha City, Hunan Province. During the incubation experiment, the minimum temperature was 20 °C and the maximum was 34 °C with an average range of 24∼30 °C. After 2, 7, 14, 21 and 28 days, soil pH was directly determined and a subsample was taken from each pot to measure the amount of DTPA extractable Cd (Lindsay and Norvell [1978](#page-6-0)) and DOC in the soil (Jones and Willett [2006](#page-6-0)). For the subsamples tested at the end of the incubation (day 28), the Cd fractions present in the soil were determined with Bureau Commune de Reference (BCR) sequential extraction method (Mossop and Davidson [2003\)](#page-6-0).

At the end of the incubation experiment, chemical fertilisers were applied to each pot at an amount equivalent to 4.5 g N per pot as urea, 1.5 g P per pot as  $KH_2PO_4$  and 4.5 g K per pot as  $KH_2PO_4$  and KCl, mixed with the rest of the soil thoroughly as a basal fertiliser. After the application of chemical fertilisers, the soil mixtures in the pots were covered with 3 cm of water above the soil surface for 2 days, and on 1 July 2011, 30-day-old seedlings of rice named Fengyuanyou 299 (Oryza sativa L.) were transplanted into the pots with a density of three seedlings per pot. The pots were kept outside with a natural day/night regimen and watered as required. Urea was applied as a top dressing fertiliser at the amount of 1 g N per pot for all treatments on 10 July 2011, and the rice was harvested on 5 November 2011. Then, the rest of the soil from each pot was homogeneously mixed, and Chinese cabbage (Brassica chinensis) seeds were planted with a density of ten seeds per pot on 8 March 2012. The pots were kept outside with a natural day/night regimen and watered as required. Five seedlings were reserved and the rest were removed from each pot 2 weeks after the Chinese cabbage was planted. Then, urea

was applied as a top dressing fertiliser in the amount of  $2 \text{ g N}$ per pot on 22 March 2012; the Chinese cabbage was subsequently harvested on 8 May 2012. After harvesting, rice grain was oven-dried at 60 °C and the weight of per pot was recorded. To determine Cd concentrations, the oven-dried (at 60 °C) rice grain, rice straw and Chinese cabbage samples were ground to pass through a sieve of 0.3 mm.

#### Analysis

The DTPA extractable Cd (water extractable Cd) in Cdcontaminated rice straw was measured with 5.0 g dried Cdcontaminated rice straw suspended in 50 mL DTPA solution (Lindsay and Norvell [1978](#page-6-0)) (deionised water) and shaken for 2 h at 20 °C. The suspension was filtered at 0.45 μm porosity with a cellulose nitrate filter. The DTPA extractable Cd in soil was measured using DTPA extraction method modified by Lindsay and Norvell ([1978](#page-6-0)). The four soil Cd fractions were sequentially extracted as exchangeable + water and acid soluble (AE), reducible (Red), oxidisable (Oxi) and residual (Res) using BCR sequential extraction method modified by Mossop and Davidson [\(2003\)](#page-6-0). The Cd concentrations in the solutions were determined by inductively coupled plasma optical emission spectrometer (ICP-OES; Varian, America).

Organic C and total N in the soil were measured by dry combustion in a CN auto-analyser (Vario MAX C/N, Germany), and total P was measured by the NaOH fusion method (Olsen and Somers [1982](#page-6-0)), respectively. Soil clay content was determined with the hydrometer method (Gee and Bauder [1986\)](#page-6-0). Soil DOC content was measured using KCl extraction method modified by Jones and Willett [\(2006\)](#page-6-0). To determine the total concentration in soil and plant materials (rice straw, rice grain, and Chinese cabbage), the samples were digested using mixtures of aqua regia-HClO<sub>4</sub> and HNO<sub>3</sub>-HClO4 (open system), respectively. Concentrations of Cd in the digested solutions were determined by ICP-OES for the total content of Cd. Three certified reference materials (CRMs), GBW070011 Chinese soil samples, three spikes and three blanks were used for quality control. Soil pH was directly determined using a pH meter (PHS-3C, Shanghai Dapu Instrument, P.R. China).

Data were analysed using one-way ANOVA tests with LSD tests to separate means. Differences were considered significant at  $p<0.05$ . Data were processed using SPSS 11.5 (SPSS, Chicago, USA) software.

# **Results**

Changes in soil pH and DOC contents

During the 28-day incubation period, the soil pH of the control treatment (R0) gradually increased from 4.72 to 6.25 (Fig. [1a\)](#page-3-0).

The pH of the soils treated with Cd-contaminated rice straw sharply increased over the first 7 days by 0.5 to 1.0 U, and it increased slightly during the remainder of the period. A significant  $(p<0.01)$  increase in soil pH was observed with the addition of Cd-contaminated rice straw during the first 3 weeks. However, the soil pH of the six treatments was between 6.20 and 6.35, and there was no significant  $(p>0.05)$  difference in the soil pH among treatments at the end of the incubation experiment (the 28th day).

The content of soil DOC in R0 remained essentially constant, at approximately 160 mg C kg<sup>-1</sup> soil, during the 28-day incubation period (Fig. [1b](#page-3-0)). By day 2, the soil DOC contents in treatments with Cd-contaminated rice straw added at the rates of 1, 2, 3, 4 and 5 % (R1, R2, R3, R4 and R5, respectively) significantly  $(p<0.01)$  increased 0.4-, 1.7-, 3.3-, 4.4and 6.4-fold, respectively, compared with the value in R0 (144 mg C kg<sup>-1</sup> soil). Following the initial increase in the first 2 days, the contents of soil DOC in R2, R3, R4 and R5 decreased from 392 to 247, 621 to 286, 781 to 373 and 1,068 to 372 mg C kg<sup>-1</sup> soil, respectively, between day 2 and 28. However, no significant decline occurred in R1.

Changes in soil DTPA extractable Cd, total Cd and Cd speciation

During the 28-day incubation period, the DTPA extractable Cd in the soils was also determined (Fig. [2\)](#page-3-0). The amount of DTPA extractable Cd in the control soil (R0) remained generally constant at approximately 0.15 mg kg<sup>-1</sup> soil during the incubation period. The addition of Cd-contaminated rice straw at rates of 1, 2, 3, 4 and 5 % significantly increased DTPA extractable Cd by 27, 69, 97, 123 and 147 %, respectively, compared with the R0 treatment. Moreover, during the 28-day incubation period, the amount of DTPA extractable Cd in Cdcontaminated rice-straw-treated soils was essentially constant.

At the end of the incubation experiment, the total Cd contents and speciation of Cd in the soils were determined using the BCR sequential extraction method (Fig. [3](#page-4-0)). The addition of Cd-contaminated rice straw at rates of 1, 2, 3, 4 and 5 % significantly increased total soil Cd by 18, 34, 48, 58 and 77 %, respectively, compared with the R0 treatment. Moreover, the Cd-contaminated rice straw incorporation resulted in significant changes in Cd distribution in the soils. Without the addition of Cd-contaminated rice straw, the most abundant fraction was the residual fraction, which occupied approximately 43 % of the total Cd after 28 days of incubation. Without the Cd-contaminated rice straw incorporation, the different fractions of Cd contributed to the total Cd content in the following order:  $Res > Red > AE > Ox$ . After the addition of Cd-contaminated rice straw at rates of 1, 2, 3, 4 and 5 %, the percentage of Red significantly increased by 5, 9, 9, 15 and 14 %, respectively, and the percentage of AE species significantly increased by 7, 11, 15, 17 and 18 %, respectively.

<span id="page-3-0"></span>Fig. 1 Changes in soil pH and DOC contents after applied Cd-contaminated rice straw during incubation period. a Changes of soil pH and b changes of soil DOC contents. R0 control (no Cd-contaminated rice straw applied) (white circle), R1 Cd-contaminated rice straw applied at 1 %  $(w/w,$  the same below) (white square), R2 Cdcontaminated rice straw applied at 2%(black triangle), R3 Cdcontaminated rice straw applied at 3%(black circle), R4 Cdcontaminated rice straw applied at 4%(black diamond), R5 Cd-contaminated rice straw applied at 5 % (black square). The up and down bar with each point indicates the range of the standard error of the mean, and separate bars in the line indicate the range of LSD  $(t=0.01)$  for different treatments in the same sampling time



Fig. 2 Changes in soil DTPA extractable Cd after applied Cd-contaminated rice straw during incubation period. See Fig. 1 for R0, R1, R2, R3, R4, R5 and bars



<span id="page-4-0"></span>

Fig. 3 Changes in Cd fractions in soil applied Cd-contaminated rice straw after 28-day incubation. See Fig. [1](#page-3-0) for R0, R1, R2, R3, R4 and R5. AE acetic acid extractable fraction Cd in soil (white square), Red reducible fraction Cd in soil (dotted square), Oxi oxidisable fraction Cd in soil (crosshatch-filled square), Res residual fraction Cd in soil (black square)

Relatively, the percentage of Res species significantly decreased by 11 to 27 % after Cd-contaminated rice straw was applied. While the concentration of Oxi species was essentially constant, their percentages decreased. After the Cdcontaminated rice straw was applied at 1 %, the order of the four fractions was  $Res > Red > AE > Oxi$ ; when the amount of Cd-contaminated rice straw added was over 1 %, the order of the four fractions was Red > AE > Res >Oxi.

#### Uptake of Cd by rice and Chinese cabbage

The addition of Cd-contaminated rice straw significantly decreased the rice grain yield by 53 to 79 % (Fig. 4) as compared with the R0 treatment. Equally, growth of Chinese cabbage was observed, while the weight was not recorded. Moreover, the addition of Cd-contaminated rice straw significantly affected the concentrations of Cd in plant tissues (Fig. [5](#page-5-0)). The



Fig. 4 Effect of Cd-contaminated rice straw application on rice grain yield. See Fig. [1](#page-3-0) for R0, R1, R2, R3, R4 and R5. Means and standard errors followed by the same letter are not significantly different (LSD,  $p<0.05$ 

concentrations of Cd in the rice grains and the rice straw were 0.111 and 0.112 mg  $kg^{-1}$  in the R0 treatment, respectively. The addition of Cd-contaminated rice straw significantly decreased the Cd concentrations in rice grains and rice straw by 39.3 to 57.3 and 12.8 to 70.2 %, respectively, compared to the R0 treatment. The lowest Cd concentrations in rice grains and rice straw were observed in the R5 and R3 treatments, respectively. In contrast, a significant increase in the concentration of Cd in Chinese cabbage was observed after the application of Cd-contaminated rice straw. For the R0 treatment, the concentration of Cd in Chinese cabbage was  $0.357$  mg kg<sup>-1</sup>. Addition of Cd-contaminated rice straw at rates of 1, 2, 3, 4 and 5 % significantly increased the concentration of Cd in Chinese cabbage by 13.9, 31.2, 61.0, 84.1 and 66.7 %, respectively, as compared with the R0 treatment.

# Discussion

During the 28-day incubation under flooding conditions, soil pH increased for all treatments as compared with the initial values, and the incorporation of Cd-contaminated rice straw significantly increased soil pH (except for the 28th day) compared to the control treatment. A possible explanation can be attributed to the reduction reactions under flooding conditions in acid soil; for example, denitrification and/or dissolution of iron hydroxides, consumed  $O_2$  and H<sup>+</sup> and the addition of rice straw may accelerate this change with the decomposition of rice straw (Cui et al. [2008;](#page-6-0) Zhu et al. [2012a](#page-7-0)). Moreover, the soil DOC content was enhanced by increasing rates of Cdcontaminated rice straw and gradually decreased with incubation time for rice-straw-amended soils. Similar changes in soil DOC content after the application of rice straw have also been observed by Katoh et al. [\(2005\)](#page-6-0) and Cui et al. [\(2008\)](#page-6-0). Rapid decomposition of rice straw by soil microorganisms appears to have resulted in significant production of DOC during the early stages (Katoh et al. [2005\)](#page-6-0). The DOC was also mineralised by soil microorganisms during the incubation periods of the experiment. In accord with previous studies, these results here show that acid paddy soil treated with rice straw led to an increase in soil pH and DOC content.

In the present study, incorporation of Cd-contaminated rice straw significantly increased the total Cd content of the soil. This may attribute to the high inputs of Cd in rice straw, that the total Cd content in Cd-contaminated rice straw was 6.66 mg kg<sup>-1</sup> and additional rates at 1 to 5 % mean that soil total Cd may increase by 0.07 to 0.33 mg  $kg^{-1}$ . Furthermore, the DTPA extractable, acid extractable and reducible Cd in soil were all enhanced by Cd-contaminated rice straw. However, little changes in the soil residual and oxidable Cd were observed after the application of Cd-contaminated rice straw. Similarly, the Cd-contaminated plant material addition that increased the extractable Cd concentrations in soil has

<span id="page-5-0"></span>Fig. 5 Effects of Cd-contaminated rice straw application on uptake of Cd by rice and Chinese cabbage. See Fig. [1](#page-3-0) for R0, R1, R2, R3, R4 and R5. Cd concentration in rice straw (white square), Cd concentration in rice grain (or blighted grain) (black square), and Cd concentration in Chinese cabbage (overground parts) (crosshatch-filled square). Bars indicate the standard errors. Means and standard errors followed by the same letter are not significantly different  $(LSD, p < 0.05)$ 



been also observed (Vázquez et al. [2008\)](#page-6-0). However, the mechanisms of the addition of Cd-contaminated plant materials on the changes of DTPA extractable Cd and redistribution of Cd in soil were much complex. Bai et al. [\(2013\)](#page-6-0) reported that Cd-free wheat straw application increased the soil DOC content, which may be a stronger factor than pH in increasing Cd solubility in an around neutral soil. Cui et al. [\(2008\)](#page-6-0) and Li et al. ([2014](#page-6-0)) both reported that the incorporation of Cd-free rice straw induced an increase in the soil pH, which may be one of the major reasons for reducing Cd solubility in acidic soils. Moreover, Cui et al. ([2011](#page-6-0)) found that Cdcontaminated Indian mustard leaves significantly increased exchangeable Cd in an acidic soil. These results indicated that the effects from the addition of plant materials on Cd extractability and speciation in soil depend not only on changes in soil pH and DOC but also on the concentration of Cd in plant material. In the present experiment, the DTPA extractable Cd, the acid extractable Cd and even the soil pH increased after the Cd-contaminated rice straw was applied. During the 28-day incubation, the soil DOC contents significantly decreased and the DTPA extractable Cd contents were constant in the ricestraw-treated soils. Moreover, the contents of Cd in tested rice straw were much high (6.66 mg  $kg^{-1}$ ), and about 95 % could be extracted by DTPA. Therefore, the Cd inputs from rice straw should be a stronger factor than those from Cdcontaminated rice straw addition that induced soil pH and DOC content changes on Cd extractability and speciation in this tested soil.

Although the Cd-contaminated rice straw application increased soil DTPA and acetic acid extractable Cd in soil, the Cd contents in rice grain and rice straw clearly decreased in this study. However, previous studies have widely reported positive correlations between rice uptake of Cd and both DTPA and acetic acid extractable Cd in soils (Zhu et al. [2012b\)](#page-7-0). This inconsistency could be attributed to the inhibition of rice growth after applied Cd-contaminated rice straw. Compared with the control treatment, the rice grain yield decreased by 53 to 79 % for the Cd-contaminated ricestraw-treated soils. Similarly, Bai et al. [\(2013\)](#page-6-0) found that rice growth was inhibited after wheat straw was added and that the contents and accumulation of Cd by rice plants decreased when the wheat straw addition was over 0.5 %, even the soluble Cd increased. This inhibition may be related to the immobilisation of N in the soil due to the high C/N ratio (38.7) of rice straw as well as the toxic effect of the organic acids produced during anaerobic decomposition of the added straw, even though much more nitrogen fertiliser was added in the present study as compared to Bai et al. ([2013](#page-6-0)). Moreover, equal or higher Cd concentrations in rice grain were observed in the present experiment than in rice straw for the same treatments. Commonly, the Cd content of rice straw was higher than that of rice grain; for example, in hybrid rice, the contents of Cd in the leaves and stems were 4 to 20 times more than those in polished rice (Zhao et al. [2006\)](#page-7-0). Shi et al. [\(2013](#page-6-0)) also found that the ratio of grain to straw Cd accumulation was 2.75 and 1.36 for cultivar Zhongzheyou No. 1 and 0.63 and 0.36 for cultivar J196, planted in soil with a Cd content of 0.55 and 3.05 mg  $\text{kg}^{-1}$ , respectively. The distribution of Cd in the aboveground parts of rice planted on Cd-contaminated ricestraw-treated soils could be attributed to the tested rice cultivar.

The concentration of Cd in oven-dried Chinese cabbage was among 0.357 to 0.658 mg kg<sup>-1</sup> in the present study, and the water content of Chinese cabbage was about 93.5 % referred to Rosen and Chen ([2014](#page-6-0)). Therefore, the concentration of Cd in fresh Chinese cabbage was about 0.023 to 0.043 mg kg−<sup>1</sup> , which did not excess the Chinese Food Safety Standard (0.20 mg kg−<sup>1</sup> , fresh weight). However, the concentration of Cd in Chinese cabbage significantly increased after the addition of Cd-contaminated rice straw.

<span id="page-6-0"></span>Positive correlations were also observed between Cd concentrations and both DTPA  $(r=0.739, p<0.01)$  and acetic acid  $(r=0.673, p<0.01)$  extractable Cd (the 28th data) in Chinese cabbage. Similar increases were observed in concentrations of Cd in rye grass and Thlaspi caerulescens planted in soil incorporated with Cd-contaminated T. caerulescens leaves (Perronnet et al. 2000). Thereby, the increase in Cd concentration in Chinese cabbage may be related to higher available Cd in the treated soils due to the release of Cd from Cdcontaminated rice straw. These results in here should indicate that the application of Cd-contaminated rice straw increased the availability of Cd in the soil related to the high Cd inputs.

Taking together all these findings, dispersion of Cdcontaminated rice straw to the uncontaminated soil may cause the accumulation of Cd in soil as available Cd. Hence, the input of Cd could be considered as the main factor determining the speciation and phytoavailability of Cd in uncontaminated soil rather than the changes of soil pH or DOC contents after applied Cd-contaminated rice straw. These results imply that in the Cd-contaminated paddy region, we should be vitally concerned about the management of Cd-contaminated rice straw to avoid contamination of adjacent environment. In other words, the contents of Cd in rice straw should be taken into account when rice straw was used as soil amendment. However, the uncontaminated soil was mixed with Cdcontaminated rice straw only for a short duration under incubation and pot conditions. Further research is needed to investigate and evaluate the long-term effects of Cdcontaminated rice straw incorporation on Cd availability in both uncontaminated and contaminated soils, especially at field scale. Moreover, in the Cd-contaminated paddy region, Cd-free organic materials, such as farmyard manure, should be applied for the improvement of soil organic matter because the Cd-contaminated rice straw was not suitably utilised.

# Conclusion

The incorporation of Cd-contaminated rice straw led to a significant increase in the soil pH and DOC. Meanwhile, such practice also clearly increased total Cd, DTPA extractable, acetic acid extractable and reducible Cd in soil. Further, the changes in soil Cd extractability and speciation may be attributed to the input of Cd in Cd-contaminated rice straw rather than its induced increase in soil pH and DOC content. However, the high additions of Cd-contaminated rice straw obviously inhibited the growth of rice and may result in the decrease of Cd concentrations in rice grain and rice straw. In contrast, the Cd concentrations in Chinese cabbage planted after rice harvest significantly increased. To conclude, the application of Cd-contaminated rice straw increased the availability of Cd in the soil for its high Cd inputs. Therefore, the Cd risk in Cd-contaminated rice straw should be taken into

account besides the grain products for the utilisation of Cdcontaminated paddy soils.

Acknowledgments This work was supported by the National Key Technologies R&D Program of China (2012BAD05B06), the National Natural Science Foundation of China (41101300, 41371318) and Natural Science Foundation of Hunan Province (13JJ4113).

### References

- Bai Y, Gu C, Tao T, Chen G, Shan Y (2013) Straw incorporation increases solubility and uptake of cadmium by rice plants. Acta Agr Scand B-S P 63:193–199
- Cui YS, Du X, Weng LP, Zhu YG (2008) Effects of rice straw on the speciation of cadmium (Cd) and copper (Cu) in soils. Geoderma 146:370–377
- Cui YS, Fu J, Chen XC (2011) Speciation and bioaccessibility of lead and cadmium in soil treated with metal-enriched Indian mustard leaves. J Environ Sci 23:624–632
- Gee GW, Bauder JW (1986) Particle-size analysis. In: Klute A et al (ed) Methods of soil analysis. Part 1. Physical and mineralogical method. Second ed. Madison, pp 383–412
- Jones DL, Willett VB (2006) Experimental evaluation of methods to quantify dissolved organic nitrogen (DON) and dissolved organic carbon (DOC) in soil. Soil Biol Biochem 38:991–999
- Katoh M, Murase J, Sugimoto A, Kimura M (2005) Effect of rice straw amendment on dissolved organic and inorganic carbon and cationic nutrients in percolating water from a flooded paddy soil: a microcosm experiment using 13C-enriched rice straw. Org Geochem 36: 803–811
- Li M, Tian X, Liu RZ, Chen WL, Cai P, Rong XM, Dai K, Huang QY (2014) Combined application of rice straw and fungus Penicillium chrysogenum to remediate heavy-metal-contaminated soil. Soil Sediment Contam Int J 23:328–338
- Lindsay WL, Norvell WA (1978) Development of a DTPA soil test for zinc, iron, manganese, and copper. Soil Sci Soc Am J 42:421–428
- Ministry of Environmental Protection P.R.C. and Ministry of Land and Resources P.R.C. (2014) Report on soil pollution in China. [http://www.sdpc.gov.cn/fzgggz/ncjj/zhdt/201404/t20140418\\_](http://www.sdpc.gov.cn/fzgggz/ncjj/zhdt/201404/t20140418_607888.html) [607888.html](http://www.sdpc.gov.cn/fzgggz/ncjj/zhdt/201404/t20140418_607888.html)
- Mossop KF, Davidson CM (2003) Comparison of original and modified BCR sequential extraction procedures for the fractionation of copper, iron, lead, manganese and zinc in soils and sediments. Anal Chim Acta 478:111–118
- Olsen SR, Somers LE (1982) Phosphorus. In: Page AL, Miller RH, Keene DR (eds) Methods of soil analysis, vol 2. Soil Science Society of America, Madison, pp 403–448
- Perronnet K, Schwartz C, Gerard E, Morel JL (2000) Availability of cadmium and zinc accumulated in the leaves of Thlaspi caerulescens incorporated into soil. Plant Soil 227:257–263
- Rosen V, Chen Y (2014) The influence of compost addition on heavy metal distribution between operationally defined geochemical fractions and on metal accumulation in plant. J Soils Sediments 14: 713–720
- Shi J, Pan GX, Zhang NM (2013) Effect of cadmium stress on Cd and Zn uptake and accumulation of different cultivars of hybrid rice. Acta Sci Circumst 33:2901–2910
- Vázquez S, Carpena RO, Bernal MP (2008) Contribution of heavy metals and As-loaded lupin root mineralization to the availability of the pollutants in multi-contaminated soils. Environ Pollut 152:373–379
- Wu L, Li Z, Akahane L, Liu L, Han C, Makino T, Luo Y, Christie P (2012) Effects of organic amendments on Cd, Zn and Cu

<span id="page-7-0"></span>bioavailability in soil with repeated phytoremediation by Sedum plumbizincicola. Int J Phytoremediation 14:1024–1038

- Zhang XL (2008) Physiological mechanisms of Cd hyperaccumulation by the ASME of Sedum alfredii H. and its regulation. Dissertation, Zhejiang University (In Chinese)
- Zhao BH, Zhang HX, Xi LL, Zhu QS, Yang JC (2006) Concentrations and accumulation of Cadmium different organs of hybrid rice. Chin J Rice Sci 20:306–312 (In Chinese)
- Zhu QH, Huang DY, Liu SL, Zhang WJ, Su YR, Wu JS (2005) Status and prospect s of crop straw comprehensive utilisation in hilly red soil region. Chin J Ecol 24:1482–1486 (In Chinese)
- Zhu QH, Huang DY, Zhu GX, Ge TD, Liu GS, Zhu HH, Liu SL, Zhang XN (2010) Sepiolite is recommended for the remediation of Cdcontaminated paddy soil. Acta Agric Scand Sect B Plant Soil Sci 60: 110–116
- Zhu QH, Huang DY, Liu SL, Zhou B, Luo ZC, Zhu HH (2012a) Flooding-enhanced immobilization effect of sepiolite on cadmium in paddy soil. J Soils Sediments 12:169–177
- Zhu QH, Huang DY, Liu SL, Luo ZC, Zhu HH, Zhou B, Lei M, Rao ZX, Cao XL (2012b) Assessment of single extraction methods for evaluating the immobilization effect of amendments on cadmium in contaminated acidic paddy soil. Plant Soil Environ 58:98–103