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RESEARCH ARTICLE

Cadmium accumulation, availability, and rice uptake in soils receiving long-term applications of chemical fertilizers and crop straw return

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Abstract

Fertilization and straw return have been widely adopted to maintain soil fertility and increase crop yields, but their long-term impacts on the accumulation and availability of cadmium (Cd) in paddy soils are still unconfirmed. Therefore, this study was undertaken in central China to investigate the accumulation, availability, and subsequent uptake of Cd by rice (*Oryza sativa* L.) in two adjacent field trials (P1 and P2, lasting for 10 and 12 years, respectively) under long-term straw return or in combination with chemical fertilizers. Obvious Cd accumulation, probably due to the notable Cd input from irrigation and traffic exhaust in the bulk soil (0–20 cm) of P1, was observed. The bulk soil of P2 received homogeneous straw return and chemical fertilizers, as did that of P1; however, the P2 soil almost showed Cd balance. Long-term straw return increased the portion of soil DTPA-extractable Cd to the total pool for both sites, but only P1 showed significant differences when compared to the controls. However, the highest Cd concentrations and the maximum bioconcentration factors in rice straw and grain were obtained using solo application of chemical fertilizers at both sites. Continuous additional applications of crop straw, in contrast, resulted in slightly decreased Cd uptake in rice straw, but not in grain. These findings demonstrate that neither long-term straw return nor fertilization leads directly to notable Cd accumulation, but that the promotion effects of long-term chemical fertilizer applications on Cd uptake in rice need more attention.

Keywords Long-term field trials · Cadmium · Bioavailability · Straw return · Fertilization · Bioconcentration factor

Introduction

Cadmium (Cd) has been listed as the priority inorganic soil pollutant in China and worldwide due to its toxicity to organisms and nonbiodegradability in soils (Zhao et al. 2010; Chen et al. 2018a). Cd accumulation in agricultural soils, especially those used for rice production, is one of the major environmental issues attracting growing concern (Chen et al. 2018b; Zhao et al. 2015). Rapid industrialization and urbanization in China over the past several decades has accelerated the Cd input to paddy soils through atmospheric deposition, irrigation, and usage of organic manures and agrochemicals (Hu et al. 2016a; Yi et al. 2018). Moreover, the high transferability of Cd in soil–rice systems contributes to an increasing contamination risk of Cd in rice (Zhao et al. 2010; Siebers et al. 2013; Chen et al. 2018a, c). As a result, the Cd concentration in rice grains has often been reported to exceed the limit set by the Chinese government (0.2 mg kg^{-1} DW) (Chen et al.

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2018a, b). The consumption of contaminated rice grains is a major pathway for human exposure to the highly carcinogenic Cd (Yang et al. 2017; Wang et al. 2016) and ultimately poses a potential threat to human health.

China has large amounts of straw residues derived from the continual increase in crop production (Xu et al. 2016; Yin et al. 2018). Conventional practices of dealing with the very large quantities of straw residues, such as open-air burning and throwing the straw away, are detrimental to air quality and soil fertility (Chen et al. 2017a; Yin et al. 2018). Therefore, crop straw returned to on-site fields has now been widely recommended in China for its advantages in mitigating the related environmental problems (e.g., air pollution and greenhouse gas emissions) (Li et al. 2018) and for improving soil fertility and productivity (Yang et al. 2015a; Zhu et al. 2015; Chen et al. 2017b; Zhao et al. 2018). However, crop straw commonly accumulates considerable Cd (Wang et al. 2015), and straw removal has been proven to be the primary output pathway of Cd in contaminated paddy fields (Yi et al. 2018). Meanwhile, crop straw also exhibits great potential for influencing Cd mobility and availability (Ok et al. 2011; Yang et al. 2015b; Xu et al. 2016; Tang et al. 2017). Many efforts have been made to elucidate the changes in soil Cd mobility and bioavailability after return of crop straw. The evidence has shown that the effects of crop straw return on Cd availability in soils are influenced by soil pH, soil organic carbon (SOC), availability of mineral nutrients (e.g., Ca, Zn, Fe), and Cd content in straw (Zeng et al. 2011; Wang et al. 2015). However, contradictory results have been reported. Some of the previous studies have indicated that the addition of crop straw may immobilize Cd via an increase in soil pH (Wang et al. 2015) and metal adsorption by ligands in organic matter (Mohamed et al. 2010; Xu et al. 2016; Tang et al. 2017) and in turn decrease plant Cd accumulation. Ok et al. (2011) revealed that rapeseed residue amendment decreased the easily accessible fraction of Cd by 5–14%, thereby reducing metal availability to the rice plant. Xu et al. (2016) reported that the rice straw and wheat straw addition reduced the Cd concentration in maize shoots by 69.5 and 66.9%, respectively. Zeng et al. (2019) found that the addition of crushed straw could inhibit Cd uptake by two crops in the cassava–peanut intercropping system. Other studies suggested that application of crop residues induced an increase in dissolved organic matter, thus enhancing soil Cd solubility and availability as well as the uptake of Cd by plants (Wu et al. 2012a; Bai et al. 2013). Xu et al. (2018) found that the long-term application of rice straw residue significantly increased ($p < 0.05$) the concentration of Cd in rice stem in double-cropping rice systems in southern China. In addition, the combined application of chemical fertilizers (mainly nitrogen, phosphorus, and potassium fertilizers) that compensated for the nutrient imbalance in straw is also a routine agricultural practice to obtain high crop yields and is an important source of Cd input to paddy soils (Jiao et al. 2012; Wang et al. 2014). For instance, continuous application of phosphate fertilizer has been identified as the main source of Cd in agricultural soils (Seshadri et al. 2016). Furthermore, applying nitrogen fertilizer was found to increase Cd uptake and accumulation in rice (Yang et al. 2016) and in wheat (Li et al. 2011). However, most of these studies were generally based on artificial soil Cd contamination and/or conducted using pots or in short-term field experiments. The results obtained from long-term field trials may be more convincing and meaningful for practical agricultural operations (Wu et al. 2012b). Therefore, the gap in information regarding Cd accumulation and availability at the field scale as influenced by long-term crop straw return or in combination with fertilization still needs to be investigated.

With a high population density and rapid industrial development, the Jianghan Plain, an area of major rice production located in central China, is confronting a growing risk of soil Cd pollution (Yi et al. 2011; Gu et al. 2016). Two adjacent field trials focusing on the effects of long-term straw return on soil quality, fertility, and productivity in a rice–wheat (*Triticum aestivum* L.) rotation system located on Jianghan Plain have been carried out for more than 10 years, but Cd accumulation and availability have not been examined in detail. Therefore, the aim of the present study is to shed light on the influence of long-term applications of crop straw alone or in combination with chemical fertilizers on Cd accumulation, availability, and uptake by rice plants. This research will provide valuable information for predicting Cd risk under long-term fertilization and crop straw return.

Materials and methods

Study area

The two long-term straw return fields where the trials were conducted (P1 and P2) are located in the core area of the Jianghan Plain in central China (longitude 112° 37′ E, latitude 30° 22′ N), which has a humid subtropical monsoon climate with an annual average temperature of 15.8–17.5 °C and a mean precipitation of 1250 mm concentrated from April to August. The soil is classified as an inceptisol according to the Keys to Soil Taxonomy (Soil Survey Staff 2014). The selected basic physicochemical properties of the bulk soils (0–20 cm) in the two study sites are listed in Table S1 in the Supplementary material. As illustrated in Fig. 1, P1 is close to State Road 318, which is one of the most important national highways across China. Moreover, the irrigation channel of P1 is connected with the industrial and domestic wastewater channels of a nearby town. However, P2 is at a distance from the road and is irrigated by water brought directly from a branch of Yangtze River.

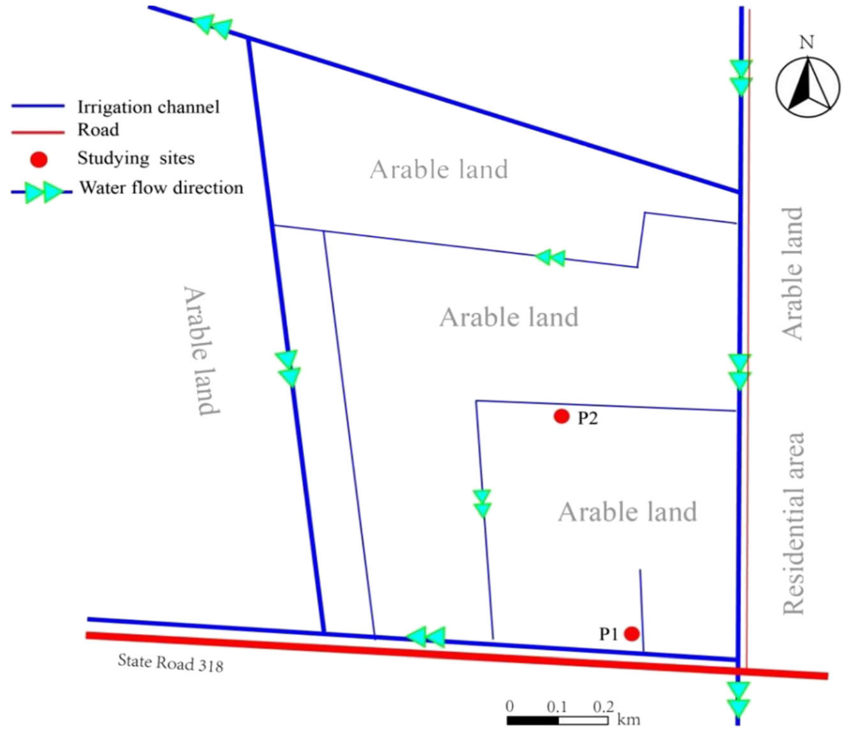

Fig. 1 Schematic diagram depicting the location and irrigation system of the two study sites

Experimental design

The three treatments of P1 consisted of CK1 (control without fertilization and straw return), NPK1 (chemical fertilizers), and NPKS1 (chemical fertilizers and straw return) since the beginning of 2007, and plots (6.0×3.5 m^2) were arranged in a randomized block design with four replicates. However, in P2, four treatments initiated in 2005, which includes S2 (straw return alone) that was added compared to those of P1, were applied to plots (5.0×4.0 m^2) arranged in a randomized block design with four replicates. Rice–wheat rotation was designed for both sites and the rice variety belonged to the *indica* hybrid cultivar. In each crop season, the crop straw was harvested from 10 cm above the ground, and the stubble and root were left in the field. The straw collected from the two sites was cut into 10 cm pieces after threshing and then mixed thoroughly, and commercially available effective microorganisms (Wuhan Heyuan Green Organism Co., Ltd., China), which were mainly composed of typical microbial communities in soils (e.g., bacteria, yeasts, fermenting fungi, and actinomycetes), were added to facilitate rapid microbial decomposition of the straw for 2 to 3 weeks. For straw returned to the plots, the wheat/rice straw was uniformly incorporated into the surface soil by tilling before rice transplantation or sowing of wheat. Nitrogen, phosphorus, and potassium were applied as urea (46% N), calcium superphosphate (12% P_2O_5), and potassium chloride (60% K_2O), respectively. Elemental analysis revealed that the Cd content in urea and calcium superphosphate was 0.01 and 0.46 mg kg^{-1}, respectively, and Cd was undetected in potassium chloride. The application rates of chemical fertilizers or straw in the corresponding treatments are shown in Table S2. There were no other additives used, except for herbicides and pesticides for weed and pest control, respectively, during both rice and wheat seasons.

Soil and rice sampling

Rice plants were harvested and separated into straw and grain at maturity in early October 2017. Plant samples, including straw and grain, were randomly collected from each plot, washed with tap water, and then rinsed with deionized water before drying to a constant weight. Straw and grain were ground with a stainless steel mill into powder.

Five subsamples were randomly collected from the surface soil (0–20 cm) in each plot after the rice harvest and were mixed thoroughly into a 1-kg sample. The compost samples were air-dried and ground to < 2 mm for pH and bioavailable metal measurements and to < 0.149 mm for the determination of SOC, total Cd, and Cd fractionation.

Chemical analyses and quality control

For total Cd determination, approximately 0.5 g finely ground plant sample was digested with 5 ml concentrated HNO_3 and

2 ml H_2O_2 (30%). Correspondingly, approximately 0.2 g finely ground soil sample was digested with 6 ml concentrated HNO_3, 2 ml HCl, and 2 ml HF in a microwave digestion system (Milestone ETHOS UP, Italy) for 1 h. After microwave digestion, Teflon digestion tubes were heated at approximately 180 °C to allow the acids to evaporate. Eventually, the digests of both plant and soil samples were diluted and filtered through 0.45-μm membranes. As the pH of soils in our study was > 7.0, and diethylenetriaminepentaacetic acid (DTPA), as described by Lindsay and Norvell (1978), was used as extractant to determine the levels of bioavailable Fe, Mn, Cu, Zn, and Cd. Briefly, 10 g soil was shaken for 2 h in 20 ml solution containing 0.005 M DTPA, 0.01 M $CaCl_2$, and 0.1 M triethanolamine (TEA) at pH 7.3. Soil Cd fractionation was carried out using a BCR sequential extraction method according to Yi et al. (2018), and metals were differentiated into an exchangeable and carbonate-bound fraction (F1), a Fe–Mn oxide–bound fraction (F2), an organically and sulfide-bound fraction (F3), and finally a residual fraction (F4). All of the digests and extracts above were stored in PVC bottles at 4 °C prior to analysis.

The Cd concentrations in the plant samples and in the paddy soils, as well as DTPA-extractable Cd concentrations, were determined by graphite furnace atomic absorption spectrometry (PinAAcle 900T, PerkinElmer, USA). DTPA-extractable Fe, Mn, Cu, and Zn concentrations were determined by atomic absorption spectrometry (PinAAcle 900T, PerkinElmer, USA). Replicate samples and certified reference materials (GBW07428 for total Cd in soil; GBW07413a for DTPA-Fe, DTPA-Mn, DTPA-Cu, DTPA-Zn, and DTPA-Cd; GBW07443 for Cd speciation; and GBW(E)100348 for Cd in plant samples) were included for quality assurance.

Soil pH was measured using a pH meter with a glass electrode at a soil to water ratio of 1:2.5 (*w*/*v*). SOC was determined by the $K_2Cr_2O_7$–H_2SO_4 method (Zhao et al. 2018). In addition, the exchangeable Ca and Mg were extracted using 1 M NH_4OAc at pH 7.0 (Bao 2000), and the concentrations were then determined by the atomic absorption spectrophotometer (PinAAcle 900T, PerkinElmer, USA).

Calculation of bioconcentration factor and translocation factor

In the present study, the bioconcentration factor (BCF) and the translocation factor (TF) are introduced. The BCF (also termed as the plant uptake factor, transfer factor) was commonly adopted to denote the transfer of heavy metals from soil to plant tissues (Chen et al. 2009; Ok et al. 2011; Ding et al. 2013; Yang et al. 2017) and is calculated using the following equation: $BCF = C_{plant\ tissues} / C_{soil}$, where $C_{plant\ tissues}$ and C_{soil} refer to Cd concentrations (mg kg^{-1}) in the plant tissues (straw or grain) and in the soil, respectively. The TF was calculated using the equation: $TF = C_{grain} / C_{straw}$, where C_{grain} and C_{straw} are the Cd concentrations (mg kg^{-1}) in grain and straw, respectively (Gao et al. 2018; Liu et al. 2018).

Statistical analysis

One-way analysis of variance (ANOVA) was performed to evaluate the significance of the treatment effects using the LSD test ($p < 0.05$). Pearson correlation analysis was carried out to explore the correlations among Cd concentrations in straw and grain and soil properties as well as bioavailable metals in soils; the data included were log_{10} transformed (except for pH) to fulfill the normality and homogeneity assumptions of variance. All of the statistical analyses were conducted using the statistical program R version 3.1.3 (R Development Core Team 2015; http://www.r-project.org/). Graphs were prepared using Origin 9.0 (OriginLab Corp, Northampton, MA, USA).

Results

Rice growth

Both straw biomass and grain yield were significantly ($p < 0.05$) enhanced in the plots receiving chemical fertilizers (e.g., NPK1 and NPK2) or the combined application of chemical fertilizers and straw (e.g., NPKS1 and NPKS2) compared to their associated but unamended control plots (Fig. 2). In particular, NPKS1 and NPKS2 had the most pronounced effects on grain yield among the treatments of P1 and P2, respectively.

Changes in soil pH, SOC, and soil bioavailable metals

A significant ($p < 0.05$) decrease in soil pH in bulk soils of NPK1 and NPK2 was observed when compared to their corresponding controls (CK1 and CK2, respectively), and additional straw return (NPKS1 and NPKS2) slightly diminished the decrease (Table 1). The SOC content significantly ($p < 0.05$) improved in plots receiving straw incorporation when compared to their corresponding controls. The DTPA-extractable Fe, Cu, and Zn and exchangeable Mg contents were significantly ($p < 0.05$) enhanced after the application of NPKS1, relative to those of CK1. In contrast, both NPK2 and NPKS2 significantly ($p < 0.05$) increased the DTPA-extractable Fe content but markedly ($p < 0.05$) decreased the DTPA-extractable Mn content in the bulk soil from P2.

Accumulation and availability of Cd in bulk soil

Except for NPKS1, which notably ($p < 0.05$) increased the DTPA-extractable Cd in bulk soil, the total and DTPA-

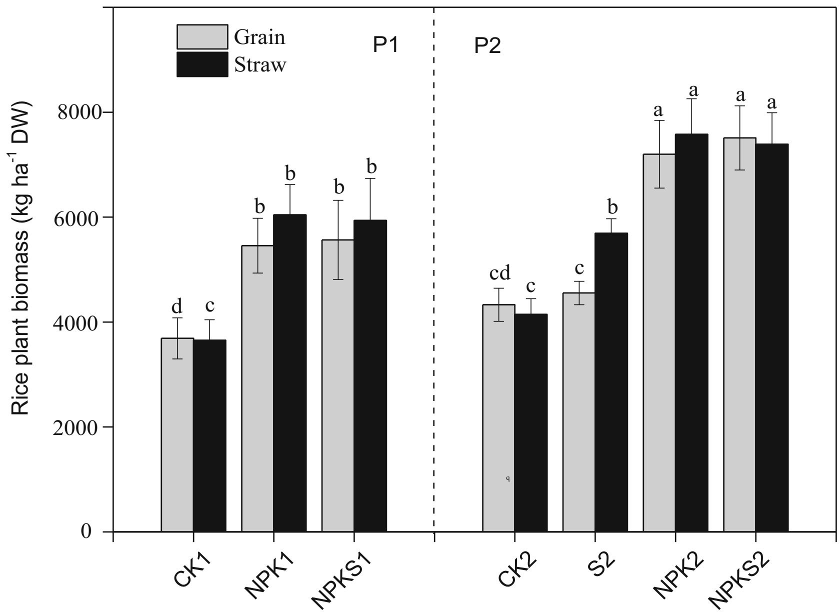

Fig. 2 Rice yield and straw biomass obtained in the different treatments at the two study sites. The treatments are as follows—P1: CK1 (control without fertilization and straw return), NPK1 (chemical fertilizers), and NPKS1 (chemical fertilizers and straw return) and P2: CK2 (control without fertilization and straw return), S2 (straw return), NPK2 (chemical fertilizers), and NPKS2 (chemical fertilizers and straw return). Values are means (±SD) of four replicates. Different letters indicate significant differences among different treatments according to LSD at the $p < 0.05$ level

extractable Cd in the rest of the treatments for both sites did not differ significantly when compared to the corresponding controls (Fig. 3). Indeed, the total Cd in NPKS1 showed a minor accumulation when compared with that in CK1, while a slight decrease of 6.9% in total Cd was observed in NPKS2 compared with CK2. The proportion of DTPA-extractable Cd in total Cd, however, showed a uniform increase after straw was incorporated at both sites, but only with significant differences ($p < 0.05$) in NPKS1, relative to CK1.

Distribution of Cd fractions

The soil Cd fraction results showed that for the bulk soil of P1, the exchangeable and carbonate-bound fraction (F1) was the highest fraction, followed by the Fe–Mn oxide–bound fraction (F2), the residual fraction (F4), and the organically and sulfide-bound fraction (F3) (Fig. 4). The percentage of F1 increased from 51.1% in CK1 to 52.5% in NPK1 and to 53.6% in NPKS1. Meanwhile, the percentage of F2 increased from 31.6% in CK1 to 32.2% in NPK1 and to 33.3% in NPKS1. Correspondingly, the proportion of F3 and F4 in NPK1 and NPKS1 decreased. Similarly, F1 was also the dominant geochemical phase in bulk soils from P2. The percentages of F2 and F3 decreased from 21.04 and 7.17% in CK2 to 18.54 and 6.78% in S2, respectively, while the percentage of F4 increased from 22.98% in CK1 to 24.39% in S2. However, it was the percentage of F1 that slightly increased from 49.73% in NPK2 to 51.46% in NPKS2, and the percentage of F4 decreased from 23.98% in NPK2 to 21.89% in NPKS2.

Cd concentrations in grain and straw

Higher concentrations of Cd in rice straw and grain were observed in P1 than in the corresponding plots in P2 (Table 2), which coincided with the results of both the total Cd and DTPA-extractable Cd contents. However, the application of chemical fertilizers alone notably ($p < 0.05$) increased the Cd concentrations in both straw and grain for the two study sites when compared with the corresponding controls. Meanwhile, BCF_{straw} and BCF_{grain} also showed an increasing trend after the application of chemical fertilizers alone, but only with a significant difference ($p < 0.05$) in P2 relative to the controls. However, the promoting effects induced by the application of chemical fertilizers alone on enhancing Cd concentrations in straw and the corresponding BCF_{straw} value were slightly mitigated after the addition of crop straw. Furthermore, the addition of crop straw caused no significant changes in Cd concentrations in grains and in the corresponding BCF_{grain} value when compared to those in plots receiving chemical fertilizer application because of the elevated $TF_{straw–grain}$ values at both sites.

In addition, there was an insignificant increase in Cd concentrations of straw and grain, under the S2 treatment, compared to those of CK2. Similarly, higher BCF_{straw} and BCF_{grain} were also obtained but did not show significant differences relative to CK2.

Table 1 Soil pH value, organic carbon content, and bioavailable metal concentration in the treatments at the two study sites

Sites	Treatments	Soil properties		Bioavailable metal concentration (mg kg^{-1})					
		pH	SOC (g kg^{-1})	DTPA-Fe	DTPA-Mn	DTPA-Cu	DTPA-Zn	Ex-Ca	Ex-Mg
P1	CK1	7.62 ± 0.08b	8.19 ± 0.31f	36.61 ± 1.60e	35.06 ± 1.20ab	2.85 ± 0.10b	0.59 ± 0.05b	4667.85 ± 168.03a	230.27 ± 25.44c
	NPK1	7.47 ± 0.04c	9.49 ± 0.11e	65.30 ± 1.76c	28.76 ± 0.97b	2.95 ± 0.11ab	0.62 ± 0.03b	4084.47 ± 253.91b	235.72 ± 37.76bc
	NPKS1	7.54 ± 0.08bc	11.18 ± 0.47d	87.01 ± 7.25b	31.52 ± 0.30b	3.21 ± 0.09a	0.87 ± 0.02a	4211.17 ± 175.87ab	264.73 ± 21.47b
P2	CK2	7.77 ± 0.08a	11.74 ± 0.49cd	52.03 ± 3.12d	36.48 ± 1.97a	3.13 ± 0.16ab	0.26 ± 0.02c	3236.13 ± 474.60c	314.59 ± 11.46a
	S2	7.76 ± 0.13a	12.97 ± 1.59ab	63.23 ± 4.22cd	39.47 ± 2.15a	3.23 ± 0.11a	0.33 ± 0.04c	2841.91 ± 302.66c	321.15 ± 5.42a
	NPK2	7.48 ± 0.07c	12.55 ± 0.15bc	121.11 ± 2.68a	25.78 ± 1.99c	3.15 ± 0.12ab	0.24 ± 0.03c	3118.37 ± 430.36c	305.73 ± 19.01a
	NPKS2	7.52 ± 0.06bc	13.80 ± 0.93a	127.26 ± 4.01a	25.27 ± 1.54c	3.19 ± 0.11a	0.25 ± 0.04c	2804.42 ± 443.22c	297.12 ± 12.98a

pH is the 1:2.5 (*w*/*v*) ratio of soil:water; DTPA-Fe, DTPA-Mn, DTPA-Cu, and DTPA-Zn mean the content of DTPA-extractable Fe, Mn, Cu, and Zn in soils; Ex-Ca and Ex-Mg represent exchangeable Ca and Mg in soils. Values are means ($n = 4$) ± SD, and different letters within columns represent significant difference at the $p < 0.05$ level (LSD test)

SOC soil organic carbon

Correlation coefficients of Cd concentrations in rice straw and grain with soil pH, SOC, and bioavailable metals in the bulk soil

Pearson correlation analysis was performed to identify the influence of pH, SOC, and soil bioavailable metals on Cd concentrations in rice straw and grain (Table 3). It can be seen that soil pH was significantly and negatively associated with the logarithmic values of Cd in rice straw and in grain in P2. In contrast, it was the SOC content that significantly and positively correlated with the logarithmic values of Cd in rice straw and in grain in P1. These results suggested that SOC and pH played a major role in influencing Cd uptake in P1 and P2, respectively.

In addition, the correlations between the logarithmic values of Cd concentrations in rice straw or grain and the logarithmic values of bioavailable metal content in bulk soil were positive at $p < 0.05$ for Fe, negative at $p < 0.05$ for Mn and Ca, and not significant for Cu, Zn, and Mg in P1. Similar results were also found in terms of Fe ($p < 0.001$), Mn ($p < 0.001$), and Ca ($p < 0.05$) in P2, where a significantly negative correlation ($p < 0.05$) between the logarithmic values of Cd concentration in straw or grain and soil Mg content was also found.

Discussion

Effects of long-term fertilization and crop straw return on Cd accumulation and bioavailability in the bulk soil

Previous studies documented that total Cd content in farmland soils was determined by both parent materials and anthropogenic activities (e.g., atmospheric deposition, irrigation, and application of chemical fertilizers and agrochemicals) (Wang et al. 2014; Yi et al. 2018). In the present study, the P1 site is close to a major national highway with heavy traffic and its irrigation channel runs through a residential area (Fig. 1), and the total Cd content at the initial sampling in 2007 in P1 was 0.49 mg kg^{-1} (Table S1) and rose to 0.63 mg kg^{-1} in control plots after the rice harvest in 2017(Fig. 3), which is higher than the risk screening values for soil contamination of agricultural land (0.6 mg kg^{-1}, pH > 7.5) set by the Chinese soil environmental quality standard GB15618-2018. It was obvious that notable Cd accumulation had occurred in bulk soils of the controls in P1, which was presumably due to the substantial amount of Cd input from the polluted irrigation water or from nearby traffic discharges. The Cd input from irrigation water could be indirectly reflected by the Cd content in irrigation channel sediments acting as a sink for Cd (Ghazban et al. 2015). One such investigation of the distribution of heavy metals in irrigation channel sediments of the same area was conducted in 2013 by Gu et al. (2016) and revealed that the

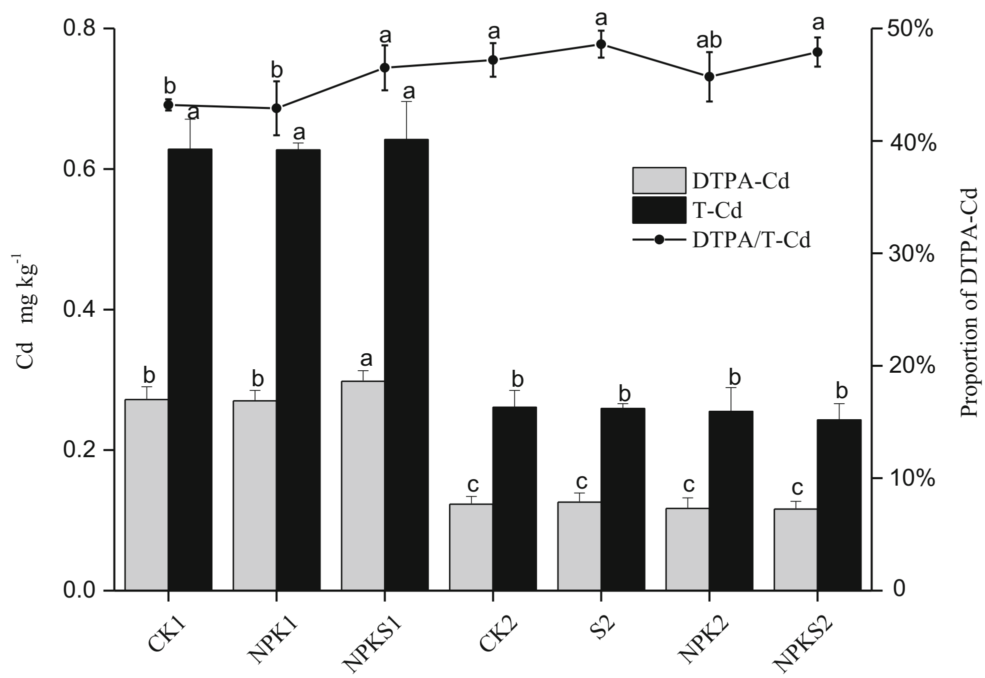

Fig. 3 Total and DTPA-extractable Cd contents and the proportion of DTPA-extractable Cd in total Cd in the different treatments at the two study sites. The treatments are as follows—P1: CK1 (control without fertilization and straw return), NPK1 (chemical fertilizers), and NPKS1 (chemical fertilizers and straw return) and P2: CK2 (control without fertilization and straw return), S2 (straw return), NPK2 (chemical fertilizers), and NPKS2 (chemical fertilizers and straw return). Values are means (±SD) of four replicates. Different letters indicate significant differences among different treatments according to LSD at the $p < 0.05$ level

mean content of total Cd for 36 sampling sites was 1.79 mg kg^{-1} (Table S1), which is significantly higher than the background value (0.17 mg kg^{-1}) and our results (Fig. 3) and implies that the irrigation water had once been polluted by Cd. In addition, higher contamination levels of Cd at the roadside sampling sites were detected during the same investigation. Therefore, more attention should be paid to the Cd input from irrigation or traffic discharges in P1. In contrast, the total Cd content in the control plots of P2 remained relatively constant at approximately 0.26 mg kg^{-1} after 12-year fertilization or straw return (Table S1 and Fig. 3). Moreover, there were no significant differences in both total Cd and DTPA-extractable Cd contents among all treatments of P2, where some of the primary input pathways of Cd (e.g., chemical fertilizers, straw return, and agrochemicals) had the same sources as those in P1. It is suggested that in the present study the application of crop straw as well as chemical fertilizers themselves did not result in a distinct accumulation risk of soil

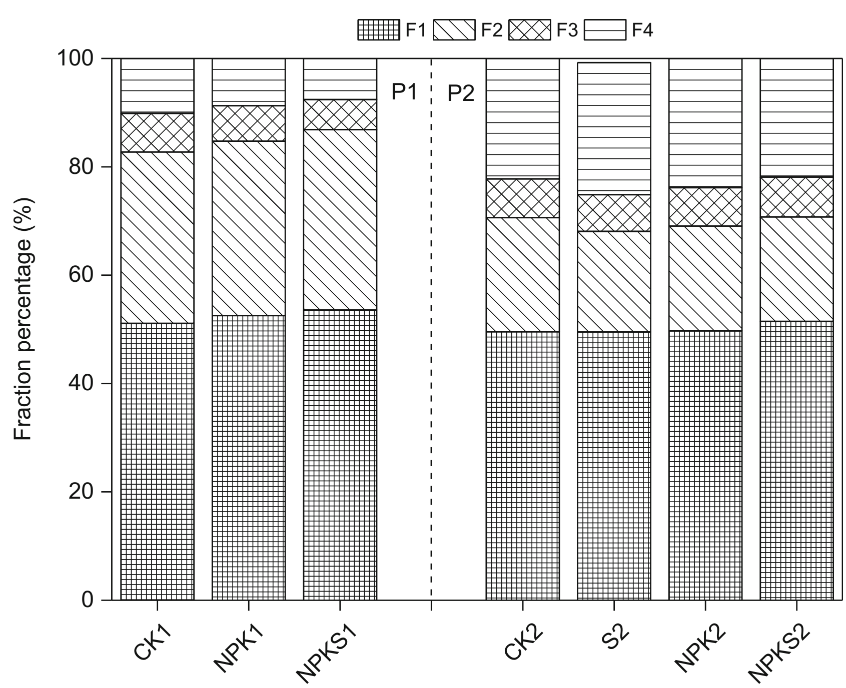

Fig. 4 Geochemical fractions of Cd in post rice soils collected from the treatments at the two study sites. The treatments are as follows—P1: CK1 (control without fertilization and straw return), NPK1 (chemical fertilizers), and NPKS1 (chemical fertilizers and straw return) and P2: CK2 (control without fertilization and straw return), S2 (straw return), NPK2 (chemical fertilizers), and NPKS2 (chemical fertilizers and straw return)

Table 2 Concentration (conc., mg kg^{-1}) of Cd in rice tissues (straw and grain) and the bioconcentration factor (BCF) and the translocation factor (TF) in the treatments at the two study sites

Sites	Treatments	Cd conc. in straw	Cd conc. in grain	BCF_{straw}	BCF_{grain}	$TF_{straw–grain}$
P1	CK1	0.17 ± 0.005b	0.034 ± 0.009b	0.272 ± 0.018bc	0.055 ± 0.018c	0.203 ± 0.062b
	NPK1	0.252 ± 0.022a	0.064 ± 0.011a	0.402 ± 0.031b	0.101 ± 0.017bc	0.251 ± 0.027b
	NPKS1	0.239 ± 0.08a	0.064 ± 0.016a	0.37 ± 0.108bc	0.098 ± 0.022bc	0.272 ± 0.022b
P2	CK2	0.06 ± 0.005c	0.021 ± 0.003b	0.231 ± 0.013c	0.079 ± 0.013bc	0.349 ± 0.072ab
	S2	0.078 ± 0.014c	0.027 ± 0.002b	0.300 ± 0.052bc	0.103 ± 0.01b	0.348 ± 0.031ab
	NPK2	0.167 ± 0.021b	0.056 ± 0.011a	0.671 ± 0.156a	0.223 ± 0.056a	0.338 ± 0.07ab
	NPKS2	0.148 ± 0.03b	0.054 ± 0.008a	0.619 ± 0.166a	0.224 ± 0.051a	0.366 ± 0.054a

BCF_{straw} and BCF_{grain} refer to the bioconcentration factor in rice straw and in grain, respectively. $TF_{straw–grain}$ refers to the value of Cd translocation factor from straw to grain. Values are means ($n = 4$) ± SD, and different letters within columns represent significant difference at the $p < 0.05$ level (LSD test)

Cd. Consistent results were found by other researchers (Wu et al. 2012b; Zhou et al. 2015; Xu et al. 2018).

Generally, the mobility and phytoavailability of Cd is associated with soil properties (e.g., pH, SOC, cation exchange capacity, and oxidation–reduction status), among which soil pH and SOC content posed more distinct effects on Cd mobility (Zhao et al. 2010; Zeng et al. 2011; Ding et al. 2013; Meng et al. 2018; Xu et al. 2018), thereby influencing the Cd uptake by rice plants (Xiao et al. 2017). In our study, notable change in soil pH occurred in plots receiving chemical fertilizers, and it was the SOC content that significantly ($p < 0.05$) increased after continuous straw return at both the P1 and P2 sites (Table 1). Our results showed that it was the soil pH, rather than SOC, that significantly ($p < 0.05$) and negatively correlated with the logarithmic values of Cd in the rice straw and in grain in P2 (Table 3), which suggests that soil pH plays a more important role in controlling Cd availability in the soils of P2 which were almost under Cd balance. However, a significant ($p < 0.05$) negative relationship between the logarithmic values of Cd concentrations in grain and the DTPA-Cd content in bulk soil was obtained in P2 (Table 3), which may be explained by the slight decrease in DTPA-extractable Cd in the bulk soil of P2 (Fig. 3) that resulted from the higher rice plant biomass and higher Cd concentrations in rice aerial tissues and, thus, higher uptake amounts of Cd from soil in the plots receiving long-term applications of chemical fertilizers. In contrast, the only significant positive relationship ($p < 0.05$) between SOC and the logarithmic values of Cd in the rice straw and grain was observed in P1, thus demonstrating a more important role for SOC in influencing Cd phytoavailability in the soils of P1 with substantial amounts of Cd input. The proportion of DTPA-extractable Cd in total Cd increased significantly ($p < 0.05$) in NPKS1 (Fig. 3), which is consistent with the elevated percentage of the exchangeable and carbonate-bound fraction and the Fe–Mn oxide–bound fraction compared with those of NPK1 (Fig. 4). Furthermore, a significant relationship ($R^2 = 0.45$, $p < 0.05$) between SOC and DTPA-extractable Cd content was observed in P1, which indicates that the role of SOC in affecting Cd availability may be ascribed to the formation of SOC–metal complexes (Liu et al. 2009; Zeng et al. 2011; Kwiatkowska-Malina 2018). Many researchers have also found that low molecular weight organic matter introduced by decomposition of straw might act as a carrier of Cd (Khokhotva and Waara 2010; Wu et al. 2012a; Bai et al. 2013; Xie et al. 2019).

Table 3 Correlation coefficients of Cd concentrations in rice straw and grain with soil pH, SOC, and exchangeable metal contents in the bulk soils at the two study sites

Sites		pH	Log_{10}^{SOC}	$Log_{10}{}^{DTPA-}_{Cd}$	$Log_{10}{}^{DTPA-}_{Fe}$	$Log_{10}{}^{DTPA-}_{Mn}$	$Log_{10}{}^{DTPA-}_{Cu}$	$Log_{10}{}^{DTPA-}_{Zn}$	$Log_{10}{}^{Ex-}_{Ca}$	$Log_{10}{}^{Ex-}_{Mg}$
P1	$Log_{10}^{Grain-Cd}$	−0.20	0.67*	0.29	0.80**	−0.61*	0.38	0.37	−0.55*	0.10
	$Log_{10}^{Straw-Cd}$	−0.38	0.52*	0.38	0.69*	−0.58*	0.34	0.19	−0.55*	0.33
P2	$Log_{10}^{Grain-Cd}$	−0.76**	0.38	−0.44*	0.90***	−0.80***	−0.13	−0.37	−0.40	−0.47*
	$Log_{10}^{Straw-Cd}$	−0.79***	0.36	−0.36	0.93***	−0.79***	0.01	−0.41*	−0.48*	−0.47*

Contents of soil bioavailable metals and Cd concentrations in rice straw and grain were log_{10} transformed to ensure homogeneity of variances

*$p < 0.05$; **$p < 0.01$; ***$p < 0.001$

Effects of long-term fertilization and crop straw return on rice growth, Cd uptake, and translocation in rice tissues

Rice growth was clearly enhanced with the addition of crop straw, let alone chemical fertilizers and their combined application when compared to the long-term nonfertilized controls at both sites (Fig. 2), which was in accordance with a number of previous studies (Hu et al. 2016b; Xu et al. 2010; Li et al. 2014; Zhang et al. 2015) and indicates that straw return is a feasible practice in terms of maintaining rice productivity.

The Cd concentrations in rice grain harvested from both P1 and P2 did not exceed the limit set by the Chinese government, which may be attributable to the alkaline characteristics of the soils in both P1 and P2. However, the application of chemical fertilizers with or without straw return was significantly ($p < 0.05$) effective in facilitating Cd uptake and accumulation in both rice straw and grain from the two sites, and the highest Cd concentrations in grain were achieved with the solo application of chemical fertilizers (Table 2); this finding is consistent with the results of Singh et al. (2010), thus demonstrating the Cd accumulation risk in rice grain under long-term application of chemical fertilizers. In contrast, additional straw incorporation resulted in higher proportions of DTPA-extractable Cd in total Cd at both sites, but with a slight decrease in Cd accumulation in rice straw. An important reason for this inconsistency is likely the competition for absorption between Cd and other mineral nutrients. As a nonessential metal ion, Cd^{2+} was absorbed and transported in rice plants by exploiting the transport systems for essential cations such as Ca^{2+}, Mn^{2+}, and Zn^{2+} which are competitors of Cd^{2+} for adsorption sites (Seshadri et al. 2016; Liu et al. 2017). The observation that the logarithmic values of DTPA-extractable Mn and exchangeable Ca showed significant negative correlations with the logarithmic values of Cd concentrations in rice straw or grain at both sites (Table 3) may confirm the above speculation. However, there was no reduction in Cd concentration in grain as was seen for rice straw after the additional straw return. This is predominantly because of the slightly increased $TF_{straw-grain}$ induced by additional straw incorporation (Table 2) which is inconsistent with the previous study of Xu et al. (2018), who found that the Cd concentrations in rice stems increased significantly but decreased significantly in rice grain under long-term combined application of rice straw and chemical fertilizers compared to that of the chemical fertilizer applications. These results may imply that the long-term straw return practices will at least not promote Cd accumulation in rice grain. However, it has been reported that transportation of Cd from root to straw and not straw to grain is likely the major process determining Cd accumulation in rice grain (Uraguchi et al. 2009; Gao et al. 2018), and the reduction of Cd enrichment in rice straw usually results in lower Cd accumulation in rice grain. Therefore, further studies should be conducted to elucidate the underlying mechanism of the elevated $TF_{straw-grain}$ after additional straw incorporation.

Conclusion

At two adjacent long-term experimental sites, the plots receiving long-term application of straw return and/or chemical fertilizers from the same sources for 10 or 12 years were under different Cd balance. Only the bulk soil at P1 showed distinct Cd accumulation, which was probably derived from irrigation and traffic discharges, and the elevated SOC content induced by straw incorporation was further beneficial for Cd accumulation. The SOC and soil pH played a major role in controlling Cd availability in the soils of P1 and P2, respectively. Despite the long-term application of straw return with or without chemical fertilizers, the proportion of DTPA-extractable Cd in total Cd increased slightly at both sites compared with untreated controls or with plots receiving chemical fertilizers; the application of chemical fertilizers was more effective in facilitating Cd uptake in rice straw and grain. Additional straw return could slightly decrease the Cd concentrations in rice straw, but not in grain, and the mechanisms involved need further investigation. Overall, long-term application of chemical fertilizers and crop straw return have no obvious impacts on Cd accumulation in the bulk soil, but we should take note of the substantial amount of Cd input from other sources and the promotion effects of long-term application of chemical fertilizers on Cd uptake in rice.

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