

EFFECT OF RICE STRAW AND ITS DECOMPOSED MATERIAL ON IMMOBILIZATION OF CADMIUM IN RED SOIL BY BIOCHAR

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ABSTRACT

To effectively utilize agricultural wastes for soil cadmium immobilization has become hot topic of environmental science. This study examined the effect of rice straw (RS), its decomposed product (DRS) and rice straw biochar (BC) on immobilization of Cadmium (Cd). Results showed that, all treatments at the 3% application rate of biochar with increasing the RS and DRS application rate from 0% to 15%, the soil pH and soil organic matter (SOM) level significantly increased. RS and DRS combined with BC in different rate made toxicity characteristics leaching procedure test (TCLP) cadmium levels in soil reduced by the percentage of 6.2-25.1 and 6.2-31.7, respectively. For European Community Bureau of Reference (BCR) sequential extraction procedure, the exchangeable-Cd and reducible-Cd fractions were transferred to oxidisable and residual-Cd fractions in both RS and DRS treatments when their combination with BC in various ratios. Overall, different rates of RS and DRS can promote biochar effect of cadmium immobilization in red soil. The most effective treatment for immobilization of cadmium by biochar is 3% for raw rice straw and 0.6% for decomposed rice straw.

Keywords: biochar, rice straw, cadmium, immobilization.

1. INTRODUCTION

Cadmium is one of the most toxic metals. Cadmium in soil, mainly coming from human activities, is easily uptaken by crops and threat to human health through the food chain (Hédiji et al., 2015; Wang et al., 2011; Xiong et al., 2016). Cadmium in soil is difficult to be degraded by microorganisms or chemical activity (Bolan et al., 2013). To reduce its soluble and exchangeable fraction in soil, chemical immobilization remediation is a potential method, which can satisfy the cultivated land resource utilization and ensure the demand for food supply (Hu et al., 2017).

Global annual output of agricultural waste is about 500 million tones (Duku et al, 2011), and its returning to the farmland soil directly is one of the most economical and realistic methods for improving soil fertility (Zhu et al., 2010). In addition, few studies reported that crop straw may decrease the available concentrations of heavy metals in soil through adsorption. Soil incorporation with 1% rice straw could significantly reduce available Cd, while addition 1% wheat straw clearly decreased Pb level in soil (Xu et al., 2016). In a similar line, Cui et al. (2008) reported that free Cd²⁺ concentration in soil decreased when adding rice straw at the rate of 6%. However, decomposition process of crop straw in soil may cause the change to immobilization

effect on heavy metal. The concentration of CaCl₂-extractable Cd from soils amended with rice straw decreased on day 4 of incubation, while clearly higher on day 7 or 21, but became comparable to the control on day 81 (Tang et al., 2017).

Crop straw biochar as immobilization material has been proven that effectively enhanced the adsorption of heavy metal and significantly reduced their mobility and phytoavailability (Park et al., 2011; Puga et al., 2015), it is also advantages in greenhouse gas emission reduction (Awad et al., 2018). Short-term pot experiment result showed that concentration of Cd in red soil decreased in toxicity characteristics leaching test (TCLP) by 42.9 and 36.7% for rice straw biochar and maize stover derived biochar, respectively at 3% application rate (Saqib et al., 2018). Similarly, the result for two years experiment revealed a substantial reduction in the exchangeable fraction of Pb and Cu by 57.56% and 54.18% respectively when adding 5% of rice straw biochar to co-contaminated soil, meaning soluble form of heavy metals transformed into insoluble forms (Abdus et al., 2019). However, the amount of biochar should be controlled below 5% to avoid the negative impact on soil properties (Matovic, 2011).

The aim of this study was to investigate the effect of rice straw and its decomposed product combined with rice straw biochar in various ratios on Cd immobilization, providing the theoretical

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evidence for remediation soil contamination by cadmium.

2. MATERIALS AND METHODS

2.1. Soil and amendments

Soil sample characterized as red soil were collected from 0-20 cm depth at Huangshi city, Hubei province, China. Soil samples were air dried, ground and sieved through 2 mm before incubation. Soil texture was measured by pipette method. The soil contained 3.2% sand, 76% silt, and 20.8% clay and was classified as silty loam soil according to the USDA classification (Klute 1986).

Rice straw obtained from a farmland of Huazhong Agriculture University, Hubei province, China, was washed with tap water, air dried and then ground to pass through 0.25 mm meshes. From the chopped rice straw, decomposed rice straw was made by using a decomposing agent purchased from Hunan Taigu Bio Technology Co., Ltd, matured 16 days; Rice straw biochar was produced at 500°C pyrolysis in an automated muffle furnace for 2 h. Selected physico-chemical properties of the soil and amendments are listed in Table 1.

2.2. Soil incubation with rice straw materials

Table 1. Basic soil and amendments properties.

Properties	Soil	BC	RS
pH	6.1	10.4	6.4
OC (g kg ⁻¹)	7.0	335.5	699.6
Total Cd (mg kg ⁻¹)	6.0	Nd*	Nd
Ash (%)		52.0	
Yield (%)		39.9	
C %		34.7	18.4
N %		0.7	0.8

Nd: Not determined*

Table 2. Design of experiment treatments.

Treatments	Biochar and rice straw application rate		Treatments	Biochar and decomposed rice straw application rate	
	BC	RS		BC	DRS
	(%)	(%)		(%)	(%)
CK	0	0	CK	0	0
BC	3	0	BC	3	0
RS-A	3	0.6	DRS-A	3	0.6
RS-B	3	0.75	DRS-B	3	0.75
RS-C	3	1	DRS-C	3	1
RS-D	3	1.5	DRS-D	3	1.5
RS-E	3	3	DRS-E	3	3
RS-F	3	6	DRS-F	3	6
RS-G	3	9	DRS-G	3	9
RS-H	3	12	DRS-H	3	12
RS-I	3	15	DRS-I	3	15

Various dose of materials (rice straw and its decomposed product) was combined with 3% biochar (w/w). Each type of material was arranged with 11 treatments, included control. Each treatment was conducted in triplicates. The experiment design was as Table 2. Polythene cups was filled with 100 g of air-dried soil. The experiment soil was homogenously amended with biochar and rice straw or decomposed rice straw

in different ratios, respectively, on dried soil basis. All treatments were kept 60% field water holding capacity during incubation, and regulate water content every 3 days. After incubation for 55 d, the soil samples were collected and ground to pass through a series of sieves for chemical properties analysis.

2.3. Chemical analysis

The pH of biochar and rice straws were measured at a 1:20 (w/v) ratio in water after stirring for 30 min, and the soil pH was measured with soil to water ratio at 1:2.5 (w/v) by an automated pH meter. The potassium dichromate volumetric method was used to measure soil organic carbon. The total Cd in soil, biochar and rice straw was determined with atomic absorption spectrophotometer (AAS-240FS mode), after digestion by mixed acids (HNO₃- HCl - HClO₄ at ratio (v/v) of 3:1:1). All methods followed Lu (1999). The solubility product of Cd was estimated from each experimental pot by toxicity characteristic leaching procedure (TCLP) following USEPA (1992). Briefly, 1.0 g of ground soil was extracted with 20 mL of 0.1M glacial acetic acid solution (pH 2.88 ± 0.02) for 18 h, then solution was filtrated and its Cd concentration was analyzed by AAS. Cadmium proportions of the samples was determined by BCR sequential extraction technique (Rauret et al., 1999).

2.4. Statistical analysis

Microsoft Office Excel 2013 was used for calculation of means and standard deviations, while Analysis of Variance (ANOVA) and Duncan Multiple Range test (p < 0.05) was used for the mean significant differences of all treatments by SPSS 20.0 software.

3. RESULTS AND DISCUSSION

3.1. Soil pH and organic carbon after amendments added

Figure 1 (A) shows that, the pH values of all treatments significantly (P < 0.05) increased compared with the control. The value of soil pH is 6.1 in CK treatment, and the highest pH value 7.1 was found in

RS-H treatment, total increased 1.0 unit. At the same application rate with 3% of biochar, soil pH values gradually increase with the raising of rice straw addition rate. It was not obviously different when application rate of rice straw ranged from 0.6% to 1.5%; enhanced from 6.6 at 1.5% application rate of rice straw treatment to 7.0 in the treatment at 6% addition dose of rice straw treatment; and there is not markedly fluctuation after incorporation of 6% rice straw application rate.

Compared to the control, soil pH value clearly increased by 0.2-1.3 units when biochar combined with decomposed rice straw in different ratios (Figure 1 (B)). Soil pH value significantly enhanced from 6.3 for BC to 6.6 for DRS-A (soil treated with 0.6% of decomposed rice straw application rate). There was not significantly change of soil pH due to application rate of decomposed rice straw at 0.6%-1%. Incorporation of decomposed rice straw ranged from 1% to 12% caused soil pH considerably raised. The highest soil pH 7.4 was found after treated with 12% and 15% decomposed rice straw application rate.

These results confirmed that rice straw and its decomposition product could promote the improvement of soil pH value by biochar.

Soil organic matter levels were significantly (P < 0.05) increased when biochar combined with rice straw in various ratios treatments compared to the control (Figure 2 (A)). In general, soil organic matter contents increased with the increasing of rice straw application rate. Soil organic matter levels ranged from 7.0 g kg⁻¹ in the control treatments to 18.2 g kg⁻¹ in RS-I (15% rice straw addition rate treatment).

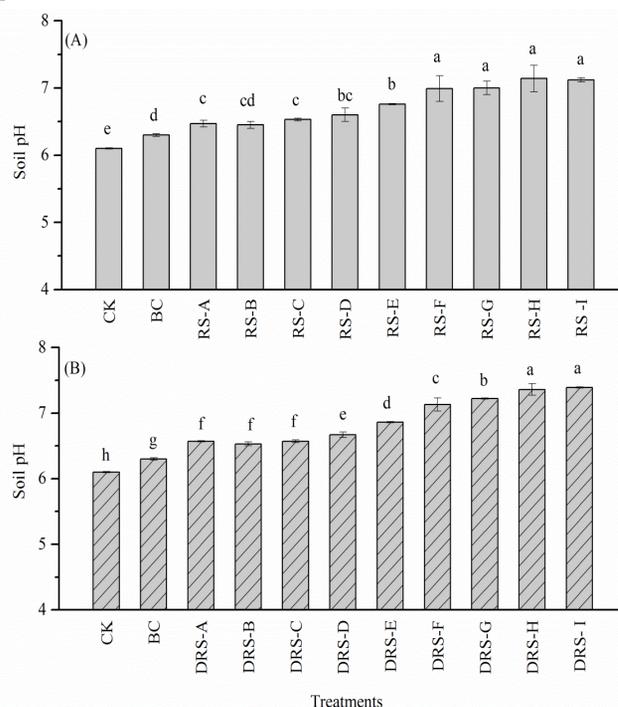


Figure 1. Soil pH change after adding rice straw and its decomposed material to treatments with biochar.

The same increased trend of soil organic matter levels was appeared when biochar was mixed with decomposed rice straw treatments (Figure 2(B)). Compared to the control, soil organic matter contents significantly increased by 1.3-2.6 times.

In general, soil organic matter increased with the increasing of decomposed rice straw. The highest amount of soil organic matter levels in soil was found in DRS-H (12% decomposed rice straw adding rate) to be 17.9 g kg⁻¹.

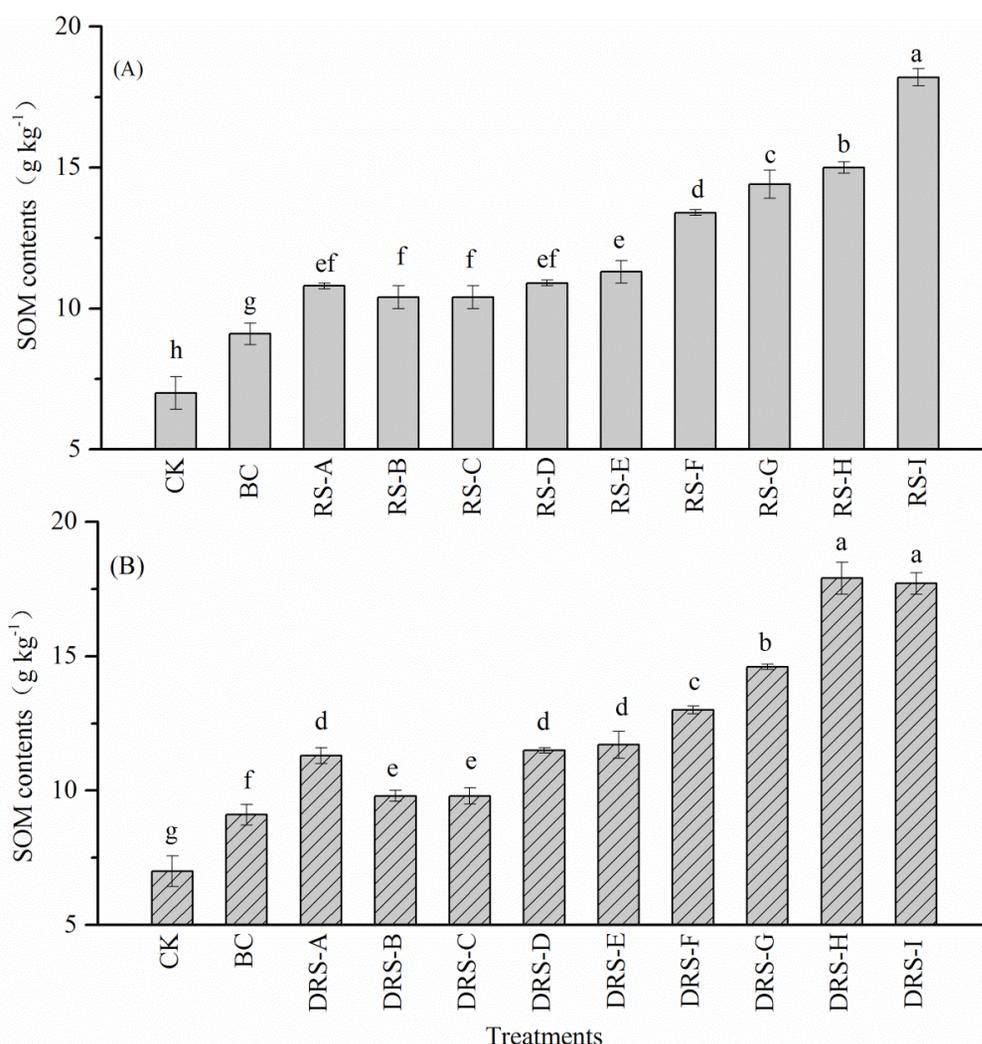


Figure 2. Soil organic matter (SOM) levels change when rice straw and it decomposed material combined with biochar in various ratios.

3.2. Mobile cadmium in soil after amendments added

Compared to the control, the amount of TCLP extractable-Cd was significantly decreased when rice straw combined with biochar in different ratios (Figure 3(A)). TCLP extractable-Cd level in CK treatment is 2.43 mg kg⁻¹. Addition of rice straw at the rate below 1% did not cause markedly change in amount of TCLP extractable-Cd. For other treatments, the decrements of TCLP extractable-Cd levels became more obvious with raising rice straw application rates. The reduction effect on TCLP extractable-Cd levels could be ordered by : 9.5% (RS-D) < 14.4% (RS-E) < 17.7% (RS-F) <

20.9% (RS-G) < 23.5% (RS-H) < 25.1% (RS-I) compared to the control. These results indicated that 1% application rate of rice straw did not change the effect on immobilization of cadmium by biochar, while application rate > 1% could improve the immobilization of cadmium by biochar. Among of all treatments, the amount of 3% rice straw application rate treatment, showed the promotion efficiency on immobilization of cadmium by biochar per unit rice straw was 4.8%, which showed the highest value. It indicated that the most effective utilization on immobilization of cadmium by biochar when the rice straw application rate is 3%.

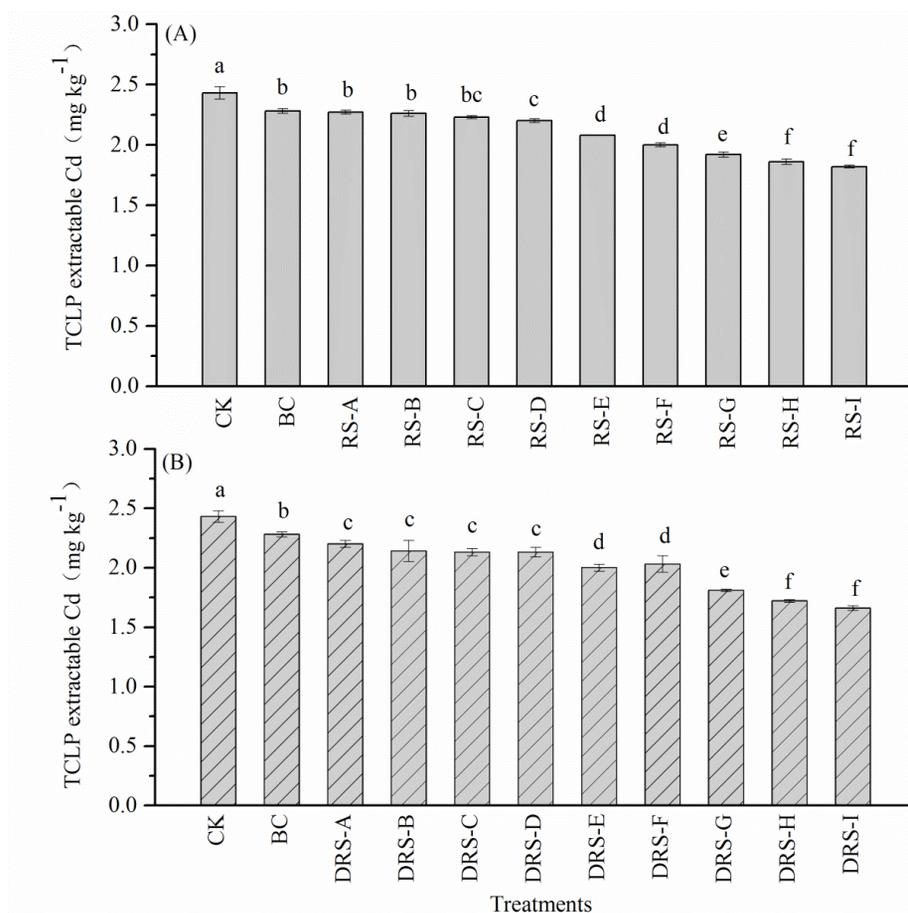


Figure 3. Effect of mixing rice straw, its decomposed material and biochar on mobile Cd level in the soil.

At 3% biochar application rate, incorporation of decomposed rice straw in different doses markedly reduced the leachability of Cd in soil (Figure 3(B)), as compared to the control. TCLP extractable-Cd was 2.43 mg kg⁻¹ in the control reduced to a minimum value of 1.66 mg kg⁻¹ in treatment DR-I, total reduced 31.7%. There are significant decline of TCLP extractable-Cd between BC and DRS-A, DRS-D and DRS-E, DRS-F and DRS-G, DRS-G and DRS-H treatments by 3.5%, 6.1%, 10.8% and 5.0% respectively. It indicated that incorporation of decomposed rice straw ranged from 0.6% to 15% application rate could gradually facilitate the reduction of TCLP-Cd effect in soil by biochar. Adding 0.6% of decomposed rice straw rate showed the promotion efficiency on immobilization of cadmium by biochar per unit decomposed rice straw was 5.8%, it was also the highest among all treatments with decomposed rice straw. It means that, 0.6% of decomposed rice straw application rate was the best choice due to the most effective utilization on immobilization of cadmium by biochar. The result also showed that the promotion effect

of 0.6% decomposed of rice straw adding rate on immobilization of cadmium by biochar is higher than the rate at 3% of rice straw, so the effect on immobilization of Cd in soil when mixing biochar and decomposed rice straw might be better than adding rice straw treatments.

3.3. Forms of cadmium in soil after treated with biochar and rice straw in various ratios

The distribution of cadmium in different fractions varies when mixing biochar and rice straw in various ratios as shown in Figure 4. In the basic of 3% biochar incorporation, the proportion of exchangeable-Cd fraction significantly decreased, responded to the application of rice straw in the range from 0% (BC) to 15% application rate by 6.0-21.9%, as compared to the control. The exchangeable-Cd was not obviously changed with incorporation below 1.5% of rice straw application rate, and markedly declined with the increasing of rice straw addition doses range from 3% to 15% .

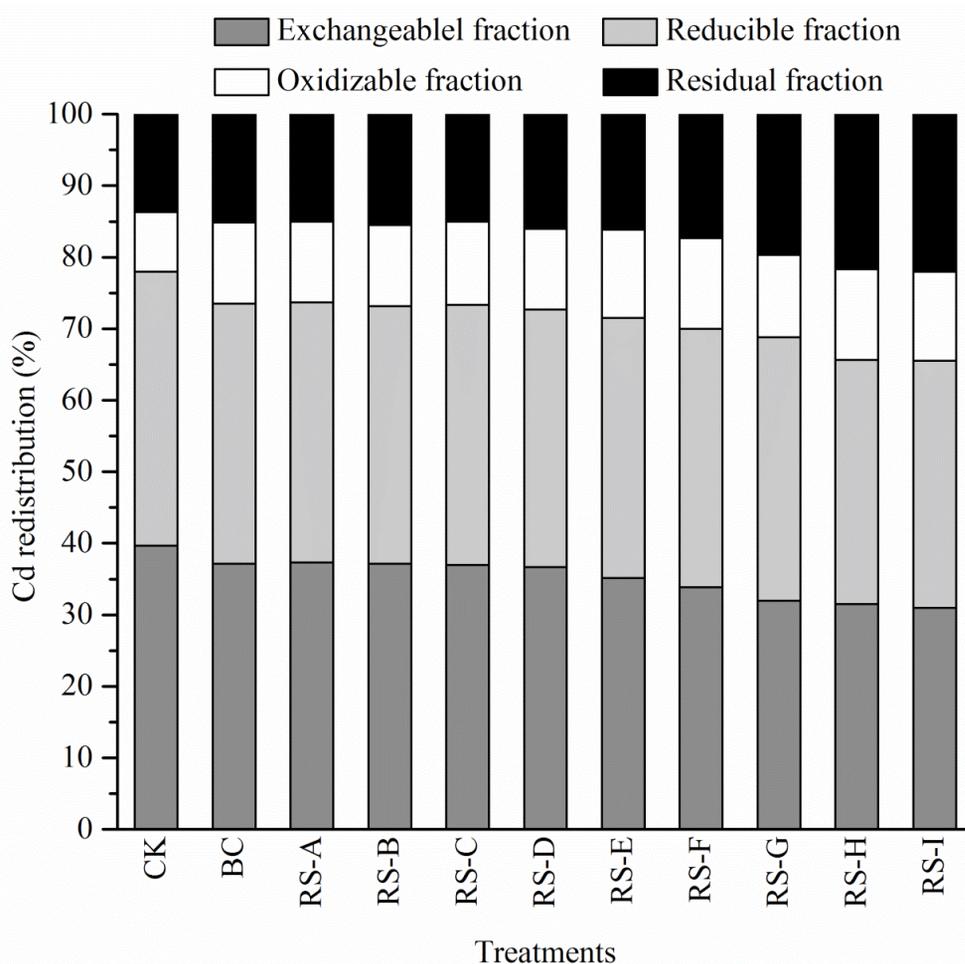


Figure 4. Effect of mixing rice straw and biochar in different ratios on Cd fractionations.

Compared to the control, the reducible fraction of Cd was reduced by 3.8-10.8% while the oxidizable-Cd fraction was increased up to 36.5-52.6% when biochar combined with rice straw in different ratios.

The upward trend of the residual-Cd fraction was found when biochar was mixed with rice straw in various ratios. Compared to the control, the proportion of residual form of Cd increased up to 9.5-60.6% in mixing biochar and rice straw treatments. Residual-Cd fraction showed little change between treatments with low rate (3%) of rice straw addition, while it was improved obviously with the increments of rice straw addition rate in other treatments, i.e. 26.5% (RS-F) < 43.6% (RS-G) < 58.2% (RS-H) < 60.6% (RS-I).

BCR sequential extraction results showed that, the exchangeable-Cd fraction and reducible-Cd fraction were transformed to the oxidizable-Cd fraction and residual-Cd fraction when biochar was combined with rice straw in different ratios, and the effect was more obvious in high application

rate of rice straw. It indicated that the soluble cadmium might be converted to insoluble after treated with biochar and rice straw in different ratios, and the higher addition rate of rice straw treatments showed the better transfer effect on different cadmium form in soil.

3.4. Cd forms in soil after treated with biochar and decomposed rice straw in different ratios

Figure 5 showed the distribution of different Cd fraction after treated with biochar and decomposed rice straw in various ratios. It indicated that the exchangeable-Cd fraction reduced from 39.7% for CK to 37.2% for BC and declined with the increments of decomposed rice straw adding rate. The lowest proportion of exchangeable-Cd fraction was found in DRS-I (at 15% decomposed rice straw application rate) to be 29.0%.

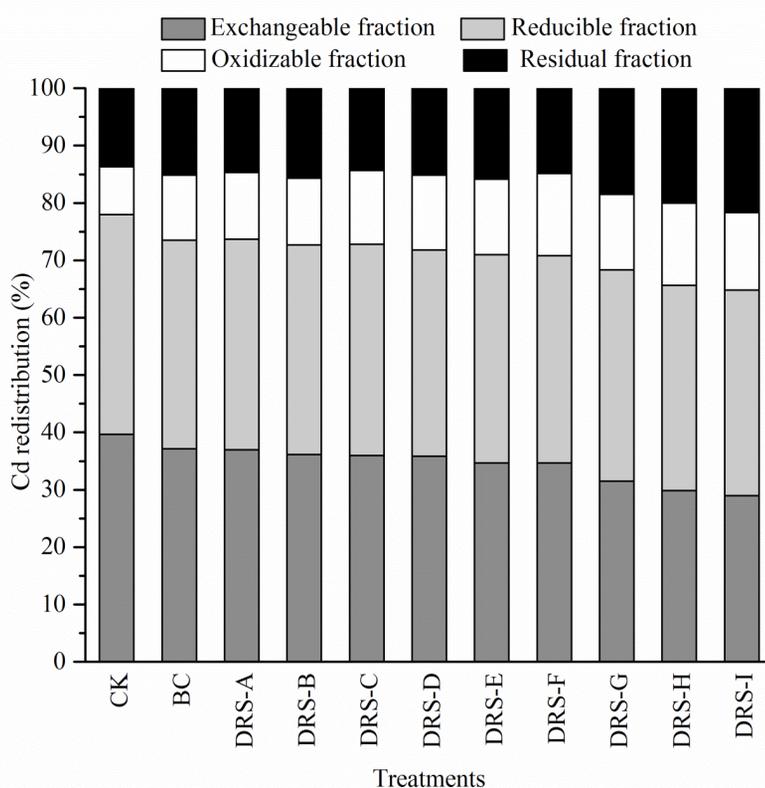


Figure 5. Effect on cadmium proportions when adding different ratios of decomposed rice straw and biochar.

Compared to the control, the reducible-Cd fraction decreased by 5.1% in BC treatment. It was remained at the same level between treatments of different ratios of biochar and decomposed rice straw.

The biggest change amount appeared in oxidization Cd fraction. The oxidization-Cd fraction increased up to 36.5-72.7% in treatments of mixing biochar and decomposed rice straw, compared with the control. Except the obvious change of proportion of oxidization- Cd fraction from CK to DRS-A and from DRS-C to DRS-D by 36.5% and 10% respectively, other treatments showed little change between each other.

Residual Cd fraction increased by 4.9-58.5% when biochar mixing with decomposed rice straw in different ratios. A slight fluctuation trend was found for amount of residual Cd when adding decomposed rice straw rate below 9%, and sharply raised with the increasing of decomposed rice straw rate with other treatments.

Mixing biochar and decomposed rice straw in various ratios could significantly vary among four fractions of Cd. The distribution of various Cd fractions in the amended soils were substantially altered by transforming the available form of Cd (exchangeable) into more stable forms (oxidization and residual). Adding decomposed rice straw

could promote the effect on immobilization of Cd in soil by biochar, and the effect also depended on the amount of decomposed rice straw. It was more obvious on immobilization of Cd in soil by biochar with the higher amount of decomposed rice straw application rate.

3.5. Discussion

In the present study, rice straw biochar effectively reduced mobile Cd in soil, which was in line with previous reports (Abbas et al., 2017; Lu et al., 2016; Yin et al., 2017). Rice straw biochar has porous structure, large specific surface area, higher alkalinity and plenty of surface organic function groups (Bian et al., 2013). Thus, it can effect on immobilization of Cd in soil through electrostatic, cation exchange, precipitation, the complexing function of oxygen function group and π -electrons mechanisms (Saqib et al., 2018; Uchimiya et al., 2014). Otherwise, rice straw biochar can also affect soil physicochemical properties and then influence Cd solubility and availability in soil, such as increase of pH, CEC, OM, EC levels in contribution to reduce of metal solubility (Khalid et al., 2017).

Our results showed that all treatments with 3% biochar application rate, the TCLP-extractable Cd decreased with the increasing of rice straw and decomposed rice straw addition rate, while

the amount of oxidizable-Cd and residual-Cd fractions in soil increased. These demonstrated that incorporation of two material (rice straw and its decomposed product) could promote the effect on immobilization of Cd in soil by biochar, and caused the amount of soluble Cd (exchangeable) convert to stable form (oxidization and residual fraction). The significant changes in soil pH and organic matter after application of rice straw and its decomposed material in various doses might influence the soil Cd stabilization by biochar. In our present research, soil pH and soil organic matter increased with the increasing of rice straw and decomposed rice straw application rate treatments. A study by Mohamed et al. (2010) showed that the variation of pH from 6.0 to 6.6, and OM from 29.8 to 46.2 g kg⁻¹ after incorporation with rice straw caused the binding constant of Cd increased from 0.6 to 3.5 mg kg⁻¹ for humic substances, thus effectively immobilized Cd in soil.

Soil pH is important for immobilization of Cd (Wang et al., 2009). The increase of soil pH declined mobile Cd content in soil (Liu et al., 2009). In this study, TCLP-extractable Cd level in soil has a significant negative correlation with soil pH. The regression equations are $Y = 5.8722 - 0.5614 \text{ pH}$ (n=11, R² = 0.966) and $Y = 5.6289 - 0.5273 \text{ pH}$ (n=11, R² = 0.925) (in the formula, Y represents TCLP-extractable Cd content in soil), respectively, when rice straw and its decomposed product combined with biochar in various ratios. After application of rice straw and decomposed rice straw, the increase of soil pH led to (1) the increasing of biochar surface variable charge and enhancement of Cd adsorption capacity, (2) fall of competition of H⁺ with Cd in soil by decreasing of H⁺ concentration in soil, contributed to the decline mobile cadmium process, (3) increase Cd hydrolysis which eventually lead to Cd hydroxides precipitation.

Similarly, our results showed that the Cd level extracted by TCLP was significantly related to soil organic matter. The regression equation are $Y = 2.8534 - 0.0614 \text{ OM}$ (n=11, R² = 0.913) and $Y = 2.8528 - 0.0664 \text{ OM}$ (n=11, R² = 0.923) (in the formula, Y represents content of TCLP-extractable Cd), respectively, when rice straw and its decomposed material combined with biochar in different ratios. These indicated that mobile Cd content decreased with increasing of organic matter amount in soil. Abundant surface active functional groups of humic substances could provide binding sites for heavy metal (Rocha et al., 2008; Yang et al., 2015; Xu et al., 2016), thus reducing their mobility in soil. Increasing of soil OM contents via the addition of rice straw governed the fixation of Cd by humic substances fractions (Mohamed et al., 2010). In addition, humic substances are also negatively charged colloid, which can adsorb Cd in soil. Above theory might also explain the increase of oxidizable-Cd fraction in the treatments when rice straw and decomposed rice straw combined with biochar.

4. CONCLUSION

Incorporation of rice straw biochar has the ability to reduce Cd mobility in red soil. Application of rice straw and its decomposed product can promote the effect on immobilization of cadmium in soil by biochar, the higher adding rate, the more significant effect. The increasing of soil pH and soil organic matter might have an effect on immobilization of Cd. But regarding on the most effective utilization on immobilization of Cd by biochar of raw material (rice straw and decomposed rice straw), it is recommended the addition rate of rice straw is 3% and 0.6% for decomposed rice straw. This confirms that rice straw and its decomposed material are beneficial for promotion efficiency on immobilization of cadmium by biochar.

ẢNH HƯỞNG CỦA ROM RẠ VÀ SẢN PHẨM Ủ CỦA NÓ ĐẾN SỰ CỐ ĐỊNH CADMIUM TRONG ĐẤT ĐỎ BẰNG THAN SINH HỌC

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TÓM TẮT

Lợi dụng có hiệu quả các phế phụ phẩm trong nông nghiệp để cố định cadmium trong đất đã trở thành một chủ đề được khoa học môi trường quan tâm. Nghiên cứu này khảo sát ảnh hưởng của rom rạ (RS), sản phẩm ủ của nó (DRS) và than sinh học làm từ rom rạ (BC) đến sự cố định cadmium (Cd) trong đất. Kết quả của nghiên cứu cho thấy, tất cả các công thức thí nghiệm trên nền xử lý với 3% than sinh học kết hợp với RS hoặc DRS với tỷ lệ từ 0% đến 15% đã làm tăng pH đất và hàm lượng chất hữu cơ trong đất (SOM) ở mức có ý nghĩa thống kê so với đối chứng. Khi RS hoặc DRS kết hợp với BC ở các tỷ lệ khác nhau đã làm cho hàm lượng cadmium trong quy trình chiết độc tính (TCLP) giảm với tỷ lệ phân biệt là 6,2 – 25,1% và 6,2 – 31,7%. Đối với quy trình chiết của Châu Âu (BCR), ở các công thức kết hợp RS hoặc DRS với BC ở các tỷ lệ khác nhau thì cadmium ở dạng ion trao đổi trong đất, cadmium ở dạng cadmi oxit trong đất được chuyển sang cadmium ở dạng phức chất với khối tử là các chất hữu cơ trong đất và cadmium ở dạng liên kết với các phức chất bền trong đất. Kết luận rằng, xử lý kết hợp với các mức RS hoặc DRS khác nhau có thể tăng cường hiệu quả cố định cadmium trong đất đỏ bằng than sinh học. Trong đó công thức xử lý kết hợp 3% rom rạ trước khi ủ và công thức xử lý với 0,6% rom rạ sau khi ủ, là những công thức làm tăng cường tác dụng cố định Cd của than sinh học tốt nhất.

Từ khóa: Than sinh học, rom rạ, cadmium, cố định.

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