



## Management of heavy metals in rice (*Oryza sativa*) soils by silicon rich biochar materials

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### ABSTRACT

Multiple heavy metals have contaminated soils with a combination of ecological consequences that make soil remediation more challenging. An experiment was conducted during 2022–23 at University of Agriculture and Forestry, Hue University, Hue city, Vietnam to evaluate the potential of silicon rich biochar from rice (*Oryza sativa* L.) husk and peanut shell in the remediation of heavy metals (Cd, Pb, Cu and Zn) present in rice soils of central Vietnam. A total of 20 samples of rice soil were taken from two distinct locations, Quang Tho commune and Thuy Phuong ward, Thua Thien Hue province, Central Vietnam to measure the quantity of heavy metals and evaluate the level of pollution. Silicon is a beneficial element and its external application as fertilizer seems impractical. Therefore, in this study, the effects of different silicon-rich materials [rice husk biochar (RHB) and peanut shell biochar (PSB)] at 6 different rates (0, 1, 2, 3, 4 and 5%) were determined in reducing heavy metal (Cd and Pb). The mean concentrations of Cd, Pb, Cu and Zn in soil samples ranged between 0.56–22.14 mg/kg; 19.48–81.30 mg/kg; 23.26–48.54 mg/kg and 28.47–55.12 mg/kg, respectively. Cd and Pb toxicity in rice soil samples was greater in Thuy Phuong ward than the average shale values. Considering the pollution load index (PLI), a total of 6 sites in Thuy Phuong ward had values >1.0 indicating pollution load in the respective sites, and Cd, Pb were the major contaminants in soils of the study area. The addition of silicon-rich materials decreased the contents of Cd and Pb in rice soils with adsorption efficiency from 22.83–38.54% and 30.69–31.53% in rice husk biochar (RHB); 20.47–29.55% and 26.77–27.87% in peanut shell biochar (PSB), respectively. Thus, RHB could be more effective to remediate soils contaminated with heavy metals when compared to other silicon-rich materials.

**Keywords:** Amendment, Heavy metal, PLI, Rice soil, Silicon

Soil contamination is primarily caused by heavy metals such as Cd, Pb, Cu and Zn. Heavy metals can enter in humans and animals through food chains, posing a serious health risk and degrading soil quality (Wijayawardena *et al.* 2016). The use of silicon (Si) in agricultural production is often cited as a method to reduce the toxicity of heavy metals (Hussain *et al.* 2015). A potential strategy for the restoration of heavy metals-contaminated areas is to use biochar as an adsorbent, which can effectively remove Cd and Pb pollutants from soil (Penido *et al.* 2019). According to several studies (Huang *et al.* 2018, Vu *et al.* 2022), interactions between minerals found in biochar and heavy metals may be responsible for up to 90% of metal removal. Soil-available silicon (Si) has been shown to reduce the availability and toxicity of heavy metals to plants (Huang *et al.* 2018). Rice husk biochar (RHB) and peanut shell biochar (PSB) are rich in silicon

and often found as agricultural wastes. Both biochar types have exceptional porosity, and with a substantial surface area improves soil fertility and decrease the bioavailability of heavy metals in soils (Huang *et al.* 2018, Leksungnoen *et al.* 2019). Amorphous silicon, most source favoured by plants, is abundant in RHB and PSB (Adrees *et al.* 2015). Additionally, RHB and PSB can raise soil pH and the activity of plant antioxidant enzymes while reducing the effects of metal toxicity on plants (Kiran and Prasad 2019). Hence, biochar rich in Si can be promising for the remediation of soil contaminated with Cd and Pb. Therefore, this study evaluated the potential of silicon rich biochar from rice husk and peanut shell in the remediation of heavy metals (Cd, Pb, Cu and Zn) present in rice (*Oryza sativa* L.) soils of central Vietnam.

### MATERIALS AND METHODS

*Study site:* Two sites representing paddy rice production as Quang Tho commune (16°32'19"N, 107°31'29"E), Quang Dien district (unpolluted soil) and Thuy Phuong ward (16°24'17" N, 107°38'7" E), Huong Thuy town (polluted

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soil) were selected for soil samples in Thua Thien Hue province, Central Vietnam. Paddy rice soils from these areas were classified as Alluvial soil (Fluvisols) (Truong *et al.* 2023).

**Sample collection:** Soil samples from these two representing sites were collected on January 2023 before the direct sowing of rice. The 20 rice soil samples were collected, 10 from each Quang Tho commune and Thuy Phuong ward sites of Thua Thien Hue province, Central Vietnam. In rice-growing fields, soil samples were collected at a depth of 0–20 cm. Each sample was rapidly packed in sealed polythene bags after sampling. A 2 mm sieve was used to separate sub-samples of the material that were air-dried first and then oven-dried at 50°C for 24 h (Saffari 2018). Each of these collected soil samples were characterized in triplicate for Cd, Pb, Cu and Zn levels.

**Experimental design:** The soil sample from each study site with the highest Cd and Pb values was selected for laboratory experiments during 2023 at University of Agriculture and Forestry, Hue University, Hue city, Vietnam. A two factors experiment arranged in a factorial design with 12 combinations per site was designed as follows: two amendments (rice husk biochar-RHB and peanut shell biochar-PSB) and six rates of amendments, Control (without amendment), 1, 2, 3, 4 and 5%. Each amendment type with application rates was added to soils and mixed in ceramic pots (10 cm × 10 cm) filled with 200 g of soil each. The pots were added with deionized water until saturation, kept for 3 days and air dried at room temperature. After that, 5 g of mixture in each pot was collected for analysis of Cd and Pb concentrations.

**Measuring the concentration of heavy metals in soil:** Cd, Pb, Cu, and Zn were analyzed using an atomic absorption spectrophotometer (iCE-3000 series, Thermo Scientific, USA). Prior to analysis, all the consumables underwent a 24 h soaking in diluted HNO<sub>3</sub> followed by a distilled water rinse. The Cu, Zn, Cd, and Pb were detected in respective limits of 0.8, 0.6, 0.07, and 0.10 ng/l.

**Biochar preparation and characterization:** Silicon rich materials like rice husk and peanut shell were chosen for biochar preparation. The raw material was initially air-dried, followed by overnight oven drying (80°C), grinding, and sieving to produce 2.0 mm particles. The biomass was ground into a powder and placed in a stainless-steel reactor. The biomass was then slowly pyrolyzed (10°C/min) in a muffle furnace with N<sub>2</sub> atmosphere at a flow rate of 50 ml/min holding at 500°C for 2 h. The colorimetric molybdenum blue method was used to determine the Si content (Qian *et al.* 2019) while the Cd, Pb, Cu, and Zn contents were

measured with an atomic absorption spectrophotometer. The properties of silicon-rich biochar are presented in Table 1.

**Adsorption efficiency (H):** The adsorption efficiency (H) of heavy metals onto adsorbents was determined as:

$$H (\%) = (C_0 - C_x) \times 100 / C_0$$

where H (%) is the adsorption efficiency of heavy metal; C<sub>0</sub> (mg/kg) and C<sub>x</sub> (mg/kg) indicate the initial concentration without material and the concentration of heavy metal after adding material, respectively.

**Pollution load index (PLI):** The method proposed by Haynes and Zhou (2022) was used to measure the PLI of surface soils of both sites in Thua Thien Hue province, central Vietnam. The PLI for a single site is the *n*<sup>th</sup> root of *n* number of multiplied together contamination factor (CF) values. The CF is the quotient obtained as:

$$CF = C \text{ Metal concentration} / C \text{ background concentration of the same metal}$$

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n}$$

where, *n* equals the number of contamination factors and sites, respectively.

When the level of *PLI* is found greater than 1, it indicates contamination, while less than 1 shows no contamination (Haynes and Zhou 2022). CF shows the contamination factor and *n* is the number of elements determined in the study (Hussain *et al.* 2022).

**Statistical analysis:** The two factors, viz. amendments and rates of application of amendments arranged in a factorial design from each site with 12 soil treatments per site were allocated. The collected data and their means were statistically analyzed for standard deviation (SD), analysis of variance (ANOVA) and their means were compared by Tukey's test HSD<sub>0.05</sub> using SPSS version 20.

## RESULTS AND DISCUSSION

**Heavy metals in rice soils:** The soil analysis showed that the Cd contents in Quang Tho commune ranged from 0.56 to 1.14 mg/kg, and were higher 9.5 to 14.8 times at Thuy Phuong ward than standard levels while ranging from 14.27 to 22.14 mg/kg (Table 2). There was a significant difference in Cd contents among surveyed sites at both locations (*P*<0.05). The use of industrial wastes, agricultural chemicals, wastes from ship-breaking industries, and mining are the main sources of toxic metal contamination in the surrounding environment (Kamani *et al.* 2018, Pham *et al.* 2021, Haynes and Zhou 2022). Pb in all soil samples ranged from 19.48 to 38.64 mg/kg (non-polluted soil) and from 45.67 to 81.30 mg/kg (polluted soil). According to the Vietnamese standard (MONRE 2015), Pb content in six rice soils of Thuy Phuong ward was higher than the standard (<70 mg/kg) from 3.14 to 11.30 mg/kg. The Cu content in rice soils of both study sites was low as compared with the standard. The Cu concentrations detected in the sampling areas had been reported previously in Bangladesh and most of the soil samples (47%) were within the range of the average shale value (45 mg/kg) (Hasan *et al.* 2022). The present study

Table 1 Characteristics of silicon-rich biochar prepared from rice husk and peanut shell

Amendment	Cd (mg/kg)	Pb (mg/kg)	Cu (mg/kg)	Zn (mg/kg)	Si (%)
RHB	0.02	2.56	9.28	5.23	27.22
PSB	0.03	4.72	10.41	7.21	12.66

Table 2 The heavy metal contents in rice soils of two study sites before sowing

Sample	Quang Tho commune, Quang Dien district				Thuy Phuong ward, Huong Thuy town			
	Cd (mg/kg)	Pb (mg/kg)	Cu (mg/kg)	Zn (mg/kg)	Cd (mg/kg)	Pb (mg/kg)	Cu (mg/kg)	Zn (mg/kg)
1	0.83 ±0.10 <sup>ab</sup>	21.20 ±3.09 <sup>ab</sup>	23.26 ±4.16 <sup>b</sup>	35.25 ±3.81 <sup>a</sup>	21.12 ±1.79 <sup>a</sup>	81.30 ±5.16 <sup>a</sup>	34.54 ±1.13 <sup>d</sup>	28.47 ±1.48 <sup>f</sup>
2	1.14 ±0.19 <sup>b</sup>	30.14 ±2.97 <sup>a</sup>	28.58 ±4.75 <sup>ab</sup>	33.56 ±3.02 <sup>a</sup>	22.14 ±1.78 <sup>a</sup>	75.18 ±4.05 <sup>ab</sup>	47.65 ±1.39 <sup>a</sup>	35.74 ±2.44 <sup>ef</sup>
3	0.65 ±0.15 <sup>ab</sup>	34.54 ±5.49 <sup>a</sup>	34.15 ±2.78 <sup>ab</sup>	39.51 ±1.42 <sup>a</sup>	19.31 ±0.75 <sup>ab</sup>	54.47 ±1.89 <sup>cd</sup>	24.18 ±1.29 <sup>f</sup>	46.49 ±0.93 <sup>c</sup>
4	1.05 ±0.12 <sup>ab</sup>	35.25 ±3.39 <sup>a</sup>	38.14 ±4.23 <sup>a</sup>	41.12 ±3.04 <sup>a</sup>	15.27 ±2.79 <sup>bc</sup>	45.67 ±5.50 <sup>d</sup>	48.54 ±1.44 <sup>a</sup>	35.72 ±0.98 <sup>ef</sup>
5	0.94 ±0.11 <sup>ab</sup>	35.56 ±2.14 <sup>a</sup>	30.25 ±4.15 <sup>ab</sup>	39.25 ±2.60 <sup>a</sup>	18.25 ±1.68 <sup>abc</sup>	75.23 ±4.16 <sup>ab</sup>	37.57 ±2.41 <sup>d</sup>	55.12 ±3.38 <sup>b</sup>
6	1.27 ±0.21 <sup>b</sup>	38.64 ±3.74 <sup>a</sup>	27.45 ±2.27 <sup>ab</sup>	38.24 ±3.29 <sup>a</sup>	10.25 ±0.76 <sup>d</sup>	78.65 ±0.76 <sup>ab</sup>	45.13 ±.67 <sup>ab</sup>	38.21 ±0.81 <sup>de</sup>
7	1.04 ±0.22 <sup>b</sup>	41.15 ±5.02 <sup>a</sup>	35.36 ±0.82 <sup>ab</sup>	31.47 ±2.80 <sup>a</sup>	14.27 ±0.84 <sup>cd</sup>	71.34 ±0.74 <sup>ab</sup>	38.45 ±0.48 <sup>cd</sup>	54.47 ±0.76 <sup>b</sup>
8	1.12 ±0.21 <sup>b</sup>	30.47 ±2.83 <sup>ab</sup>	34.24 ±1.46 <sup>ab</sup>	35.23 ±5.70 <sup>a</sup>	21.64 ±0.34 <sup>a</sup>	59.32 ±0.72 <sup>c</sup>	29.47 ±2.74 <sup>e</sup>	65.42 ±5.13 <sup>a</sup>
9	0.75 ±0.12 <sup>ab</sup>	19.48 ±3.76 <sup>b</sup>	30.18 ±2.96 <sup>ab</sup>	34.26 ±2.44 <sup>a</sup>	17.47 ±0.82 <sup>abc</sup>	69.45 ±0.72 <sup>b</sup>	42.41 ±2.56 <sup>bc</sup>	44.45 ±2.36 <sup>cd</sup>
10	0.56 ±0.06 <sup>a</sup>	33.25 ±2.21 <sup>a</sup>	31.05 ±3.84 <sup>ab</sup>	32.25 ±2.13 <sup>a</sup>	18.56 ±0.94 <sup>abc</sup>	73.14 ±2.15 <sup>ab</sup>	45.79 ±0.95 <sup>ab</sup>	46.54 ±0.82 <sup>c</sup>
Tukey HSD <sub>0.05</sub>	0.54	11.81	12.42	11.76	4.68	10.04	4.58	14.67

Means followed by the same letter(s) within the columns do not differ significantly,  $P < 0.05$ . Values after ± are SD.

results also confirm that the Cu concentrations were lower compared to other industrial cities in the world (Gujre *et al.* 2021). The Zn contents ranged from 31.47 to 41.12 mg/kg (Quang Tho commune) and from 28.47 to 55.12 mg/kg (Thuy Phuong ward). Likely, the Zn concentrations in river alluvium soils had been reported in a range of 66.40–78.50 mg/kg (Hasan *et al.* 2022). Similar concentrations of Zn had been reported in China where multi-heavy metals

pollution in soils includes mainly Cd, Pb, Zn which exceed the Chinese National standard by 7, 1.5 and 0.9% of soil, respectively (Wu *et al.* 2022).

*CF and PLI<sub>site</sub> of different heavy metals in rice soils:* The CF values of Cd, Pb, Cu, and Zn in each sampling unit of Quang Tho commune were less than 1, hence, the rice soils at this commune were low in pollution with metals (Table 3). Similarly, PLI<sub>site</sub> values in the Quang Tho commune were

Table 3 CF and PLI<sub>site</sub> of heavy metals in rice soils at two study sites

Sample	Quang Tho commune, Quang Dien district					Thuy Phuong ward, Huong Thuy town				
	CF				PLI <sub>site</sub>	CF				PLI <sub>site</sub>
	Cd	Pb	Cu	Zn		Cd	Pb	Cu	Zn	
1	0.69	0.30	0.23	0.18	0.30	14.08	1.16	0.35	0.14	0.95
2	0.76	0.43	0.29	0.17	0.35	14.76	1.07	0.48	0.18	1.08
3	0.43	0.49	0.34	0.20	0.35	12.87	0.78	0.24	0.23	0.87
4	0.70	0.50	0.38	0.21	0.41	10.18	0.65	0.49	0.18	0.87
5	0.75	0.51	0.30	0.20	0.39	12.17	1.07	0.38	0.28	1.08
6	0.85	0.55	0.27	0.19	0.40	6.83	1.12	0.45	0.19	0.90
7	0.69	0.59	0.35	0.16	0.39	9.51	1.02	0.38	0.27	1.00
8	0.75	0.44	0.34	0.18	0.38	14.43	0.85	0.29	0.33	1.04
9	0.75	0.28	0.30	0.17	0.36	11.65	0.99	0.42	0.22	1.02
10	0.37	0.48	0.31	0.16	0.27	12.37	1.04	0.46	0.23	1.08
Mean	0.67	0.45	0.31	0.18	0.36	11.89	0.98	0.39	0.23	1.01

below 1, indicating that the rice soils in this area were not polluted by heavy metals. In contrast, CF values of Cd and Pb in most of the samples in Thuy Phuong ward were higher than 1, especially with CF values of Cd, indicating that rice soils in this area were contaminated with Cd and Pb. PLI sites of samples 2, 5, 7, 8, 9, and 10 were higher than 1, showing the high risk of heavy metal contamination (Cd, Pb) at this site. Yadav and Yadav (2018) also indicated that the pollution load index (PLI) of soil was useful to assess the pollution level in the soil. The PLI revealed a baseline level of contamination in 28 locations from Al-Shamiyah city, Iraq (Al-Khuzai and Maulud 2022) and the PLI values for Co, Zn, Cu, and Pb also showed class 2 of local pollution from Ishaqi, Iraq (Al-Dabbas and Abdullah 2020). Similar result was found in Bangladesh where the contamination factor (CF) was higher for Cu and Cd (Hasan *et al.* 2022). As per above, the studied soils were highly contaminated by Cd and it was observed that pollution load index (PLI) values of all other heavy metals for all sampling units were not more than one (Islam *et al.* 2018).

*Absorption efficiency of Cd and Pb in rice soil:* The adsorption efficiency of Cd and Pb depends on the type and rate of amendments (Table 4). Application of RHB and PSB reduced the Cd and Pb contents with highest adsorption capacity found for RHB at 5%, with 22.8 to 38.54% for Cd (reduced 0.29–8.22 mg/kg Cd content in soil compared to the control) and 30.69 to 31.53% for Pb (reduced 11.86–25.63 mg/kg Pb in soil compared to the control) at both study sites, especially in Thuy Phuong ward (polluted soil). When RHB biochar was added, the materials itself had an exchange capacity of relatively high conversions and adsorb Cd and Pb into their structure, reducing the mobility of Cd and Pb in the soil, thus at the higher rate of biochar, the higher Cd

and Pb adsorption capacity were found (Li *et al.* 2019). Likely, a similar trend was found with PSB applications for Cd and Pb content at different rates. At different application rates, the Cd content changed significantly compared to the control soil in Thuy Phuong ward. At 4% and 5% application rates, the Cd content decreased significantly, but it was still above the allowed thresholds of the standard. Compared to the Pb adsorption capacity, the Cd adsorption capacity of the material was much higher. Nevertheless, RHB most promising soil amendment is related to Cd and Pb absorption ability. In the present study, the application of Si-rich amendments, viz. rice husk and peanut shell biochars successfully decreased the levels of heavy metals in rice soils. These findings had been reported earlier with the application of Si-rich biochar to reduce heavy metal accumulation in rice soil (Rizwan *et al.* 2018). According to Saffari (2018), rice husk biochar has a high pH, and cation exchange capacity, which further enhances the adsorption effect of surface complexation and ion exchange. As a result, RHB effectively reduced the bio-effectiveness of heavy metals in composite heavy metal-contaminated soil. Heavy metal toxicity has been observed to be reduced by silicon by lowering the exchangeable state of the metals in soil (Adrees *et al.* 2015).

The study's findings showed that several soil samples taken from Thuy Phuong ward in Huong Thuy town had average Cd and Pb concentrations greater than the accepted limit. According to PLI values, 6 out of the total 10 sites in Thuy Phuong ward had a value of more than 1, indicating this site had a high pollution load. The results also showed that the contamination factors for Cd and Pb were significantly greater than for Cu and Zn, indicating that Cd and Pb were the main pollutants in the soils of the

Table 4 Effects of Si-rich biochar on reducing heavy metals and absorption efficiency in rice soils

No.	Material type	Rate (%)	Quang Tho commune				Thuy Phuong ward			
			Cd (mg/kg)	H (%)	Pb (mg/kg)	H (%)	Cd (mg/kg)	H (%)	Pb (mg/kg)	H (%)
1 (Control)	Rice husk biochar	0	1.27 <sup>a</sup>	0.00	38.64 <sup>a</sup>	0.00	21.12 <sup>a</sup>	0.00	81.30 <sup>a</sup>	0.00
2		1	1.22 <sup>ab</sup>	3.94	34.21 <sup>a</sup>	11.46	19.21 <sup>abc</sup>	9.04	76.44 <sup>bc</sup>	5.98
3		2	1.20 <sup>abc</sup>	5.51	30.45 <sup>a</sup>	21.20	16.56 <sup>cde</sup>	21.59	70.25 <sup>d</sup>	13.59
4		3	1.14 <sup>a-d</sup>	10.24	29.25 <sup>a</sup>	24.30	15.89 <sup>def</sup>	24.76	68.47 <sup>d</sup>	15.78
5		4	1.04 <sup>bcd</sup>	18.11	28.34 <sup>a</sup>	26.66	14.54 <sup>ef</sup>	31.16	62.32 <sup>ef</sup>	23.35
6		5	0.98 <sup>d</sup>	22.83	26.78 <sup>a</sup>	30.69	12.98 <sup>f</sup>	38.54	55.67 <sup>e</sup>	31.53
7 (Control)	Peanut shell biochar	0	1.27 <sup>a</sup>	0.00	38.64 <sup>a</sup>	0.00	21.12 <sup>a</sup>	0.00	81.30 <sup>a</sup>	0.00
8		1	1.24 <sup>a</sup>	2.36	35.41 <sup>a</sup>	8.36	20.22 <sup>ab</sup>	4.26	78.96 <sup>ab</sup>	2.88
9		2	1.21 <sup>ab</sup>	4.72	33.22 <sup>a</sup>	14.03	17.96 <sup>bcd</sup>	14.96	74.56 <sup>c</sup>	8.29
10		3	1.18 <sup>abc</sup>	7.09	31.56 <sup>a</sup>	18.32	16.89 <sup>cde</sup>	20.03	69.45 <sup>d</sup>	14.58
11		4	1.11 <sup>a-d</sup>	12.60	30.12 <sup>a</sup>	22.05	15.21 <sup>def</sup>	27.98	63.26 <sup>e</sup>	22.19
12		5	1.01 <sup>cd</sup>	20.47	27.87 <sup>a</sup>	27.87	14.88 <sup>ef</sup>	29.55	59.54 <sup>f</sup>	26.77
F <sub>test</sub>			**	-	ns	-	**	-	**	-
Turkey HSD <sub>0.05</sub>			0.19	-	19.44	-	3.08	-	3.51	-

Means followed by the same letter(s) within the columns do not differ significantly,  $P < 0.05$ .



study area. Reducing the amount of heavy metals in rice soil might be done effectively by using Si-rich materials. The use of RHB at 5% was the best rate and a viable option for heavy metal remediation in agricultural fields. Hence, this study provides the basis for the use of rice husk biochar (RHB) for agricultural output waste and offers a promising source for remediation of heavy metal contaminated soils.

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