

Research Article

Application of Biochar in the Remediation of Contaminated Soil with High Concentration of Lead and Zinc

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Received 16 December 2020; Revised 11 January 2021; Accepted 20 January 2021; Published 30 January 2021

Academic Editor: bingxiang yuan

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Soil contamination in the Panjiachong lead and zinc mining areas has become a severe problem in Hunan Province, China. As the traditional stabilization technology comes with soil degradation, it is urgent to find a novel binder that is more eco-friendly. It has been proved that biochar can immobilize heavy metals, but limited research has been conducted on the contaminated soil with high concentration. In this study, 5%, 8%, and 10% biochar derived from the rice straw were used to remediate contaminated soil with high concentration of lead and zinc. Portland cement (PC) was adopted as the control group. The results showed that after 56 d curing, the biochar-treated soil had a neutral pH and EC value and higher soil fertility compared with the PC-treated soil. The results from the toxicity characteristic leaching procedure test indicated that the biochar is more effective than PC on heavy metal immobilization. Germination index (GI) value was used to evaluate the phytotoxicity of the treated soil; the GI values of treated soil with 8% and 10% biochar were both higher than 80%, while all the PC-treated groups failed to achieve this GI value, which indicated the potential revegetation is applicable for the biochar-treated soil.

1. Introduction

Mine tailings and wastewaters are created during the mining activity, which is one of the main sources of the heavy metal contaminants migration to the environment. Hunan Province, China, which has a huge population and abundant reserves of nonferrous metals at the same time, is a center of the mining industry and agriculture in central China [1]. It was reported that the Xiangjiang River, which is the water resource for drinking and irrigation, is severely polluted by mining activities [2]. Other studies indicated that the high concentration of lead (Pb), zinc (Zn), cadmium (Cd), and other heavy metals are found in the soil around the mining areas in the Xiangjiang Valley [3, 4]. Wang et al. [1] investigated the heavy metal pollution in soil as well as

vegetables in the Hunan Province and indicated that soil pollution posed a potential risk to human health and food safety. The traditional solution to heavy metal-contaminated soil is mixing cement-based binders such as Portland cement (PC) into the soil. However, this stabilization process would eventually cause degradation as well as hardening of the contaminated soil [5]. Meanwhile, the application of PC and other cement-based materials would increase soil pH to a certain value that makes the stabilized soil unacceptable by plants and microorganisms [5]. Due to those shortcomings of the cement-based materials, it is now very urgent to find a new alternative binder which is more eco-friendly.

Recent research has shown that the biochar could immobilize the heavy metals in soil, leading to a lower leaching concentration and lower plant availability of the heavy

metals [6, 7]. Unlike the cement-based material, the biochar is derived from biomass such as rice straw, rice husk, and bovine feces at a relatively low temperature (<700°C). It has been reported that as a carbon-enriched and porous material, the biochar is able to increase water and nutrient retention as well as microbial activity of soil [8–10]. Those superior characteristics of biochar make it an ideal alternative binder to immobilize heavy metals in contaminated soil. However, contaminated soil used in this study, which was obtained from a mining field in Zhuzhou, Hunan, had a very high concentration of lead (12510 mg/kg) and zinc (11927 mg/kg), while limited research has been conducted on the effectiveness of biochar when applied into contaminated soil with that high concentration pollution. The biochar can effectively change the porosity and the pore distribution in soil, which can improve the pore structure of soil and the circulation of the water and air in the pores [11, 12]. Also, some researchers focus on the engineering application of remedied soil [13, 14].

This study presented a systematic investigation on the physicochemical properties, fertility characteristics, leaching characteristics of heavy metals, and the phytotoxicities of contaminated soil before and after the treatment. Two types of binders were used in this study, i.e., biochar and PC. The biochar was self-prepared from rice straws as a new type of a binder; PC was used for comparison purposes. 5%, 8%, and 10% content of the binder were added to contaminated soil.

2. Materials and Methods

2.1. Test Materials. The contaminated soil used in this study was obtained from a mining field 60 km away from Zhuzhou City, Hunan Province, as shown in Figure 1. The Olympus DELTA handheld X-ray fluorescence (XRF) was used in the field to select the heavy metal-contaminated soil with high concentration of Pb and Zn. The selected soils were then excavated, sealed in proper bags, and transported to the laboratory. The basic properties, including the total heavy metal concentration and test methods and standards, are summarized in Table 1.

The biochar used in this study was self-prepared using the air-dried rice straws that are easy to obtain in China, and the reuse of this material would help decrease the air pollution in China caused by rice straw burning. The rice straws were thermo-decomposed at 500°C under an oxygen-limited condition for 5 hours. Nitrogen was also pumped into the thermal decomposition furnace during the heating process [21]. The biochar was then air-cooled and ground to pass a 2 mm sieve. The basic properties of the biochar and the test methods or standards are listed in Table 2. Locally produced 325# PC was used as the control binder. The contents of biochar and PC were selected as 5%, 8%, and 10% (dry weight basis), respectively.

2.2. Preparation of Specimens. The contents of biochar and PC were selected as 5%, 8%, and 10% (dry weight basis), respectively. The water content of the treated soil was selected as 25%, which is the natural water content. The

predetermined weight binders were added into the contaminated soil and mixed thoroughly using a portable electronic mixer for 10 min. Approximately 127 g of the treated soil was added to a stainless cylindrical mold with a 50 mm diameter and 100 mm height and then compacted into the stabilized materials with a 50 mm diameter and 50 mm height. The density of stabilized materials was controlled at 1.30 g/cm³ as per the requirement given by CJ/T340-2011 [19], as shown in Figure 2. The soil specimen was then extruded from the molds and sealed in a polyethylene bag and sent for curing for 56 d under the standard curing condition (20 ± 2°C, relative humidity of 95%). After 56 d standard curing, those specimens were taken out for the following tests. Totally, 24 specimens were prepared, and for each group of treated soil, there were four identical specimens.

2.3. Testing Methods. Table 3 shows the tests carried out on specimens in this study. The pH and EC value of the treated soil were measured as per the method given by ASTM D4972-13 [18] and CJ/T340-2011 [19], respectively. The soil fertility properties, including the content of alkali-hydrolyzable nitrogen (N), available phosphorus (P), available potassium (K), organic content, and cation exchange capacity (CEC), were tested according to the method recommended by CJ/T340-2011 [19].

To evaluate the immobilization effectiveness and leaching toxicity of the contaminated soil after the biochar treatment, the TCLP test was carried out according to the USEPA test method 1311 [23]. The standard of CJ/T340-2011 [19] also requires that the soil for potential revegetation reuse needs evaluating the phytotoxicity. The germination index (GI) of the treated soil was obtained in this study to investigate the difference between the biochar-treated soil and the PC-treated soil. To obtain the GI value of each group, 100 mg air-dried treated soil was mixed with the distilled water in a 1:2 ratio (weight basis) and put into a rotation device at 160 rpm for an hour, and the leachate was then collected and transferred into a Petri dish where 10 Chinese cabbage seeds were placed on a filter paper. The seeds were then cultured in darkness for 24 h with a temperature of 25°C. After the 24 h cultivation, the germination percentage for each Petri dish and the root length of each seed were measured. For each type of treated soil, there were five identical Petri dishes. There was a control group that the distilled water was added into the Petri dish instead of the treated soil leachate. The GI value was calculated by the following equation:

$$GI = \frac{(P_1 \times l_1)}{(P_0 \times l_0)} \times 100\%, \quad (1)$$

where P_1 is the germination percentage of the treated soil, l_1 is the average root length of the seeds in the treated soil group, P_0 is the germination percentage of the distilled water group, and l_0 is the average root length of the seeds in the distilled water group.

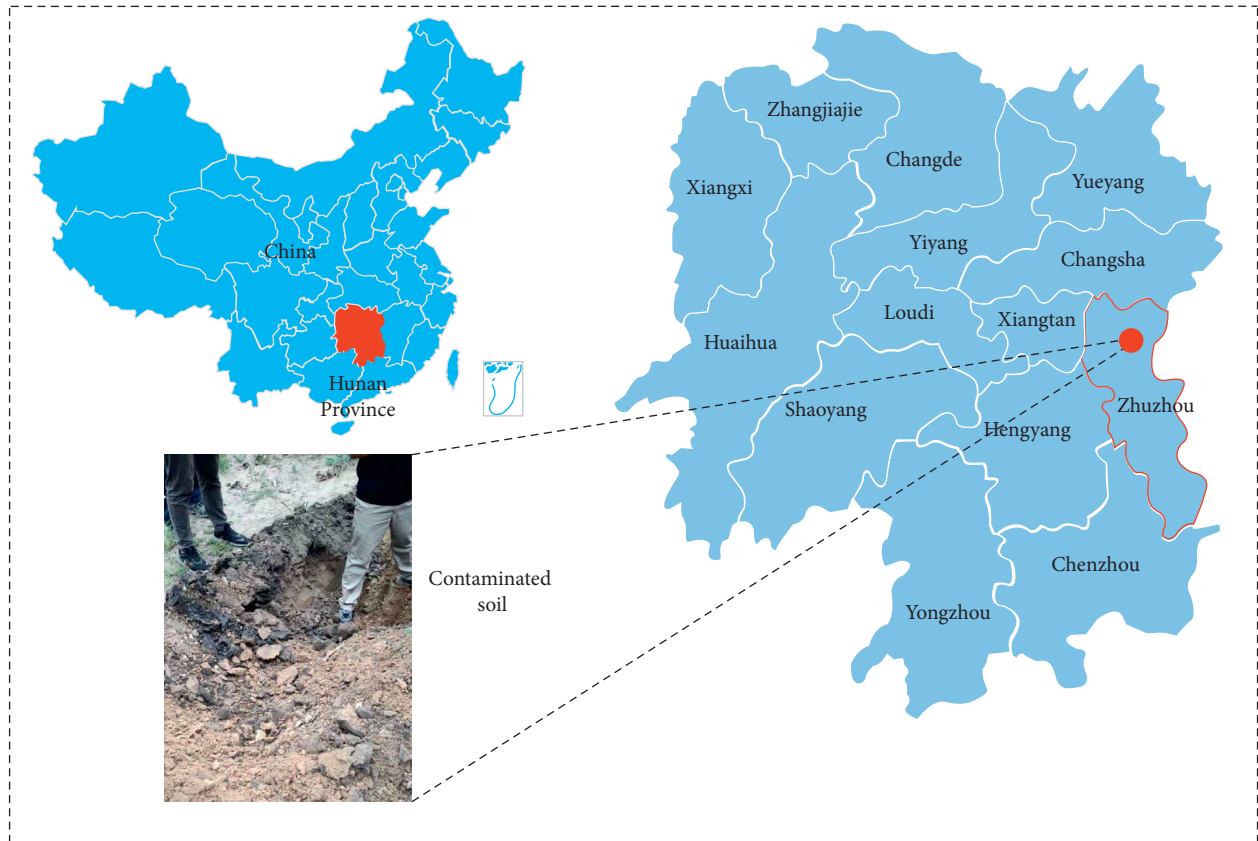


FIGURE 1: Location of Panjiachong lead-zinc mine.

TABLE 1: Basic properties of the contaminated soil.

Technical index	Value	Testing standard or method
Water content, w (%)	25	ASTM D2216-10 (ASTM 2010) [15]
Specific gravity, Gs	2.69	ASTM D5550-14 (ASTM 2014) [16]
Liquid limit, wL (%)	35.1	ASTM D4318-10 (ASTM 2010) [17]
Plastic limit, wp (%)	24.5	ASTM D4318-10 (ASTM 2010) [17]
Specific surface area (m ² /g)	41.25	Brunauer–Emmett–Teller (BET) method
pH (S/W = 1 : 1)	6.59	ASTM D4972-13 (ASTM 2013) [18]
EC (S/W = 1 : 5, ms/cm)	3.01	CJ/T340-2011 (MOHURD 2011) [19]
Contaminant concentration (mg/kg)	—	—
Pb	12510	USEPA method 3050B (USEPA 1996) [20]
Zn	11927	—

TABLE 2: Basic properties of the biochar derived from the rice straw.

Technical index	Value	Testing standard or method
pH (S/W = 1 : 1)	8.51	ASTM D4972-13 (ASTM 2013) [18]
Total C content (g/kg)	471.4	—
Total N content (g/kg)	31.3	—
Ash content (%)	6.71	ASTM D1762-84 (ASTM 2013) [22]
Specific surface area (m ² /g)	78.2	Brunauer–Emmett–Teller (BET) method
Contaminant concentration (mg/kg)	—	—
Pb	3.1	USEPA method 3050B (USEPA 1996) [20]
Zn	28	—

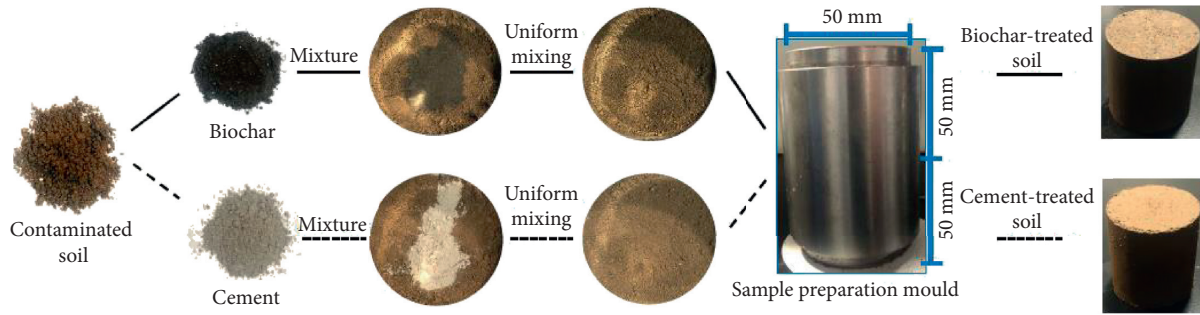


FIGURE 2: Flow chart of remediation of contaminated soil with high concentration of lead and zinc.

TABLE 3: Testing program for the contaminated soil before and after the treatment.

Binder type	Binder content (%)	Standard curing time (d)	Testing programs
Untreated	—	0	pH, EC, soil fertility (N, P, K, CEC, and organic content), TCLP, and phytotoxicity property (GI)
Biochar	5%, 8%, 10%	56	
PC			

3. Results and Analyses

3.1. Physicochemical Characteristics. Figure 3 shows the pH value of contaminated soil before and after the biochar and PC treatment. It can be seen in Figure 3 that the pH values of the treated soil increased after the treatment. It is also noticed that for both biochar and PC-treated soil, the pH value increased with the binder content. This is due to the alkalinity of the biochar and PC binder. However, according to the CJ/T340-2011 [19], the acceptable soil pH value is from 5.5 to 8.3, which means the pH values of the PC-treated soil are not suitable for applying revegetation after the soil treatment. On the other hand, it can be seen from Figure 3 that all contaminated soils after the biochar treated still have pH values ranging from 7.2 to 7.9, which are in an ideal pH interval for the potential revegetation.

Figure 4 shows the EC value of the biochar and PC-treated soil. According to Figure 4, adding biochar into the contaminated soil can significantly decrease the EC value of the soil. The EC value is used to evaluate the salinity-alkalinity of the soil, and the standard upper limit is 1.8 ms/cm [19]. It can be seen that when the biochar content is higher than 5%, the treated soils have lower EC values than the standard limit. On the contrary, the PC binder increases the soil EC value regardless of the binder content value. All the PC-treated soils have EC values ranging from 4 to 5 ms/cm, which means soils show an unacceptable salinity-alkalinity for the plants.

3.2. Soil Fertility Properties. The fertility properties, including the content of alkali-hydrolyzable nitrogen (N), available phosphorus (P), available kalium (K), organic content, and cation exchange capacity (CEC) of the untreated soil, the biochar-treated soil, and PC-treated soil, are concluded in Table 4. The standard limits in Table 4 are given by CJ/T340-2011 [19] for the revegetation evaluation. It can

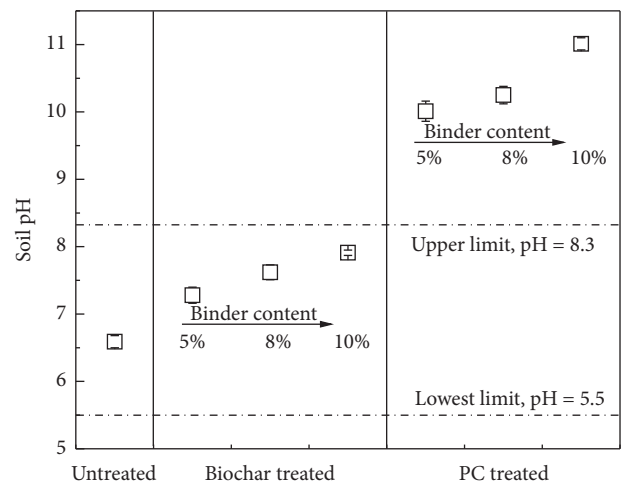


FIGURE 3: Soil pH values after the biochar and PC treatment.

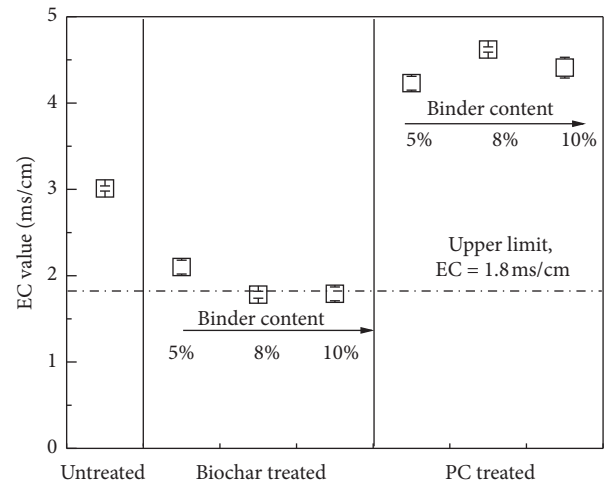


FIGURE 4: Soil EC after the biochar and PC treatment.

TABLE 4: The fertility properties of the biochar and PC-treated soils.

	Standard limit	Untreated soil	Biochar-treated soil			PC-treated soil		
			5%	8%	10%	5%	8%	10%
N (mg/kg)	≥40	52	68	84	102	41	43	38
P (mg/kg)	≥8	12.8	19.2	20.1	28.7	8.7	5.1	3.7
K (mg/kg)	≥60	79	139	154	155	51	47	50
Organic content (g/kg)	≥12	15.6	17.4	16.9	18.3	10.2	9.7	9.1
CEC (cmol (+)/kg)	≥10	5.6	12.1	17.8	19.4	5.7	6.1	3.2

be seen that the amendment of the biochar increases the fertility properties of the contaminated soil. Those results are in accordance with the reports given by Novak et al. [24]. It is also noticed that the CEC value of the biochar-treated soil increased with the content of the biochar, which is attributed to the higher surface charge found on the surface of the biochar [25]. On the other hand, adding PC into the soil does not show a positive effect on soil fertility improvement. All the relative indexes of the PC-treated soil are lower than those found in the biochar-treated soil. It is also noticed from Table 4 that when the content of PC increases to 10%, all the results do not meet the standard requirement, which indicates that the treated soil is no longer suitable for re-vegetation use.

3.3. Leaching Toxicity. The average concentration of the lead (Pb) and zinc (Zn) in the TCLP leachate of the contaminated soil before and after the immobilization is shown in Figures 5 and 6, respectively. It can be seen that the biochar-treated soil has lower Pb and Zn leaching concentration than the PC-treated soil. According to the USEPA [20, 23] and MEPC [26], the acceptable leaching concentration for Pb and Zn is 5 mg/L and 100 mg/L. It can be seen that all the soils treated by the biochar meet those requirements, while the PC-treated soils have poor immobilization effects on the contaminants. It is noticed that both leaching concentrations of Pb and Zn decreased with the increase of biochar amendment. However, there is no significant difference in the leaching toxicity between soils with 8% and 10% content of biochar, which is in accordance with the variation in CEC values shown in Table 4.

On the other hand, the leaching toxicity of PC-treated soil is apparently higher than the standard limit, which indicates that compared with biochar, the PC cannot effectively immobilize the heavy metals in the high concentration contaminated soil. This is due to the retardant effects on the hydration process of the cement-based materials caused by the presence of Pb and Zn in the contaminated soil [27].

3.4. Phytotoxicity Properties. The average GI values for each type of the treated soil obtained from the test are calculated according to equation (1), and the results are shown in Figure 7. According to the CJ/T340-2011 [19], the requirement of the GI value for the planting soil is >80%. From Figure 7, it can be seen that when amendments of biochar are

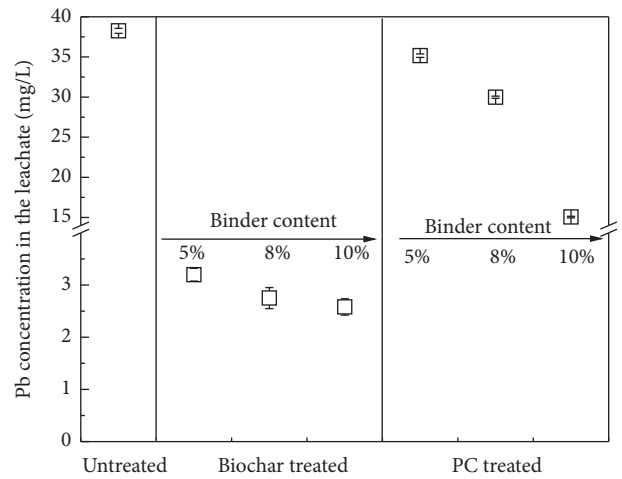


FIGURE 5: TCLP Pb leaching concentration.

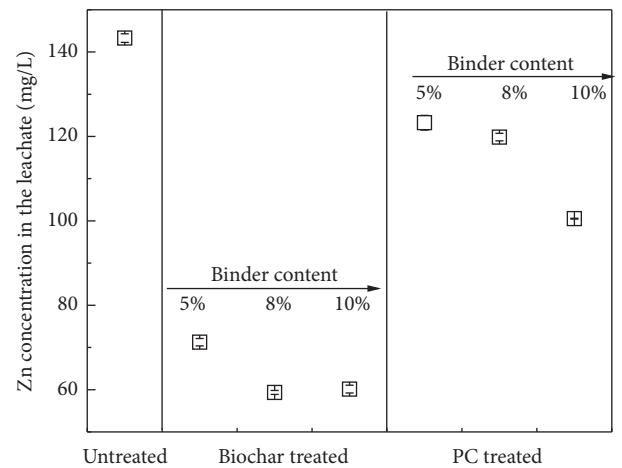


FIGURE 6: TCLP Zn leaching concentration.

8% and 10%, the treated soils have GI values higher than 80%, which meets the standard requirement given by CJ/T340-2011 [19]. These results indicated that the treated soils with 5% and 8% biochar contents are preferable for the revegetation use. The GI values of the PC-treated soils are as low as it is expected, as their higher pH and EC values shown in Figures 3 and 4 and lower soil fertility shown in Table 4 indicates that the PC-treated soil is not designed for re-vegetation use.

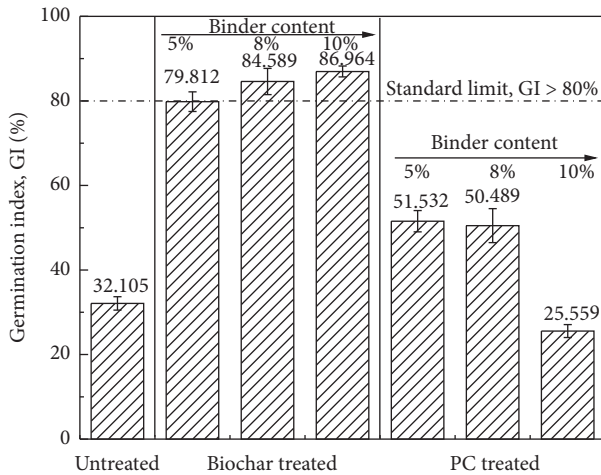


FIGURE 7: Germination index of the contaminated soil before and after the treatment.

4. Discussion

This study shows that the rice straw biochar binder compared with the PC binder has a superior reclamation effect on the soil with high concentration Pb and Zn contamination. This superior effect is reflected in terms of neutral pH value and salinity-alkalinity, higher soil fertility, lower TCLP leaching toxicity, and phytotoxicity.

According to Tang et al. [28], the electrostatic attraction is the main mechanism for the immobilization of heavy metals in the biochar-treated soil. The higher CEC values measured from the biochar-treated soil (Table 4) have positive effects on the immobilization of the Pb and Zn, as there is more negative charge on the soil surface [29]. Meanwhile, the higher available phosphorus (P) content found in the biochar-treated soil leads to the formation of insoluble heavy metal-loaded phosphate [28, 30], which contributes to the Pb and Zn immobilization. Lu et al. also indicated the hydroxyl and carboxyl functional groups could absorb the heavy metal contaminants in the soil [30]. This lower leaching toxicity of biochar-treated soil leads to less phytotoxicity shown in Figure 7. On the other hand, the superior fertility properties found in Table 4 make biochar-treated soil applicable to revegetation. The higher alkali-hydrolyzable nitrogen (N), available phosphorus (P), and available kalium (K) in the biochar-treated soil also pose positive effects on the cabbage seeds germination. Other studies also found that biochar can effectively improve plant growth by increasing nutrient availability and microbial efficiency [31, 32].

Although the results of this study show that biochar derived from the rice straw is a good alternative binder to the traditional PC due to its superior immobilization effects on the heavy metals and its improvement effects on the soil fertility properties, the phytoavailability of the contaminants contained in the biochar-treated soil and the potential heavy metal accumulation in the biosphere still need more investigations in the future. The future research can focus on

the partitioning degree of heavy metals from the easily exchangeable fraction to less bioavailable organic bound fraction and form distribution of heavy metals absorbed by plants.

5. Conclusion

This study investigates whether treated by biochar, the high concentration lead and zinc contaminated soils are suitable for the use of planting soils. Tests include soil pH and EC, fertility properties, TCLP, and phytotoxicity properties. Based on this study, the following conclusions can be drawn:

- (1) The pH of biochar and PC-treated soil increased with the binder content. Compared with PC, biochar treated the contaminated soils that have neutral pH values ranging from 7.2 to 7.9, which is suitable for the potential revegetation.
- (2) When the biochar content is higher than 5%, the treated soils have lower EC values than the standard limit (1.8 ms/cm), whereas the PC-treated soils have EC values ranging from 4 to 5 ms/cm.
- (3) Compared with PC, the amendment of the biochar increases the fertility properties of the contaminated soils. Especially, the CEC value of the biochar-treated soil increased with the content of the biochar.
- (4) Compared with PC, biochar-treated soil has lower Pb and Zn leaching concentration and meet the USEPA and MEPC acceptable leaching concentration (Pb = 5 mg/L and Zn = 100 mg/L).
- (5) Amendments of biochar are 8% and 10%, and the treated soils have GI values higher than 80%, in which the standard requirement is given by CJ/T340-2011 [19]. The treated soils with 5% and 8% biochar contents are preferable for the use of the revegetation. However, the PC-treated soil is not satisfied with the design of revegetation.

Data Availability

The datasets generated during the current study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

The authors would like to acknowledge financial support from the Xuchang University Key Scientific Research Foundation (2020ZD014), Key Research and Development and Promotion Projects in Henan Province (science and technology public relations) (212102310458), and Key Scientific Research Projects of Colleges and Universities in Henan Province (21B560015).

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