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Research Article

Evaluation of Acid Leaching on the Removal of Heavy Metals and Soil Fertility in Contaminated Soil

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Heavy metal-contaminated soils were leached with various acid reagents, and a series of treatments was assessed to understand soil fertility after acid leaching. Aqua regia digestion and a five-step sequential extraction procedure were applied to determine heavy metal distribution. The average total concentrations of Zn, Cd, Cu, and Pb for contaminated soil were 1334, 25, 263, and 525 $mg{\cdot}kg^{-1}$ based on the ICP/AES quantitative analysis. Other than Pb extracted by H_2SO_4, over 50% removal efficiency of other heavy metals was achieved. A five-step sequential extraction revealed that the bound-to-carbonate and bound-to-Fe-Mn oxides were the major forms of the heavy metals in the soil. The addition of organic manure considerably promoted soil fertility and increased soil pH after acid leaching. Seed germination experiments demonstrated that after acid leaching, the soil distinctly inhibited plant growth and the addition of manure enhanced seed germination rate from 35% to 84%. Furthermore, the procedure of soil turnover after acid leaching and manure addition greatly increased seed germination rate by 61% and shortened the initial germination time. Seed germination in untreated soil was superior to that in acid-leached soil, illustrating that the phytotoxic effect of acid leaching is more serious than that of heavy metals.

1. Introduction

Soil pollution is a serious environmental issue worldwide [1, 2]. Heavy metals are considered a serious soil pollutant because of their toxicity, persistence, and nondegradable characteristics in the environment [3–5]. In addition, the self-purification process of heavy metals present in contaminated soil is slower than that in contaminated air and water; thus, inadequate treatment of contaminated soil can pose a great human health risk. In Taiwan, more than 50,000 hectares of farmland has been contaminated by heavy metals to the fourth level classification, and more than 160 sites have been identified to be contaminated by heavy metals. Surveys state that approximately 785 hectares of farmland is contaminated by zinc to the fifth level classification [6].

The Er-Jen River located in the south western Taiwan is one of the most seriously contaminated rivers, caused by the waste from metal reclamation, smelting, and disposal of motors and electrical capacitors that released a large amount of heavy metals, polychlorinated biphenyls (PCBs), and polychlorinated dibenzo-p dioxins and dibenzofurans (PCDD/DFs) into the water [7]. Due to lack of environmental consciousness during early agricultural cultivation, the Er-Jen River was used as an irrigation source, which caused a detrimental heavy metal contamination to the paddy field. Therefore, remediation of the contaminated farmland near the Er-Jen River is of high priority.

To remediate heavy metal-contaminated soils, many effective techniques have been developed and employed, such as electrokinetics, phytoremediation, chemical extraction, vitrification, solidification, stabilization, and flotation [8–11]. However, soil washing is the most commonly used treatment technology for remediation of metal-contaminated soil. Several washing reagents, namely, water, acids, bases, chelating agents, alcohol, and other additives, have been investigated [12]. Increased attention has been recently focused on the use of citric acid or the chelating agents such as ethylenediaminetetraacetic acid

(EDTA), nitrilotriacetic acid (NTA), and diethylenetriaminepentaacetic acid (DTPA) to remove heavy metals from contaminated soils [13–15].

Although acid leaching is an effective treatment for heavy metal removal from soil, it severely damages soil fertility and its use for remediating farmland is time-consuming. The rapid enrichment of soil fertility after acid leaching is increasingly becoming a topic of concern for farmers. Therefore, this study used acid leaching techniques to remove heavy metals from contaminated farmland that can be helpful in understanding the mobility and bioavailability of metals in contaminated soil and to investigate the extraction efficiencies of heavy metals. In addition, the assessment of soil fertility and seed germination tests after the acid leaching procedure were investigated to understand the condition of remediation in the contaminated farmland.

2. Experimental

2.1. Sampling Area. Soil samples were collected from the top layer (0–30 cm) of a paddy field near the Er-Jen River in Tainan city, Taiwan province, China, which has been recognized as a previous history of irrigation with metals-rich wastewater. The paddy field contaminated with heavy metals has been strictly prohibited cultivating. Soil samples were collected by mixing ten subsamples from each site within a 50×50 meter area and were recorded for the central point position by using GPS equipment. The soil samples were air-dried at room temperature and then were ground with an agate mortar to pass through a 2 mm sieve. After sieving, the soil samples were experimentally pretreated by a vortex mixer with 100 rpm for 24 h to ensure the complete mixing of the samples.

2.2. Chemical Analysis. Soil properties, including soil pH, organic matters, soil texture, and cation exchange capacity, were determined. Particle size distribution was obtained by the pipette method after removal of carbonate, organic matters, and MnO_2. Carbonate was removed by 1 M NaOAc with a pH 5 at 60°C, and organic matters and MnO_2 were digested by 30% H_2O_2 [16]. The soil pH value was measured on a mixture of 1 : 1 soil/deionized water by a glass electrode [17]. Organic matter content was determined by the Walkley–Black wet oxidation method [18]. Cation exchange capacity was determined by the ammonium acetate method at pH = 7 [19]. Total content of heavy metals in soils was extracted by aqua regia digestion with a volume ratio of 1 : 3 HNO_3/HCl at 120°C for 8 h. The suspension was filtered through a 0.2 μm filter paper after the digestion process and was placed into precleaned 100 ml volumetric flasks with 0.5 M HNO_3 for analysis. The contents of heavy metals were analyzed by inductively coupled plasma atomic absorption spectrometry (ICP/AES). To preclude unexpected contaminations in the analysis procedure, all equipment used were washed with a phosphate-free soap and cleaned with deionized H_2O. The detection limits for metals were as follows: Cd (0.05 mg·kg^{-1}), Zn (0.02 mg·kg^{-1}), Pb (0.06 mg·kg^{-1}), and Cu (0.05 mg·kg^{-1}). The percentages of recoveries for each metal were ranged from 96.7% for Cd to 103.2% for Zn. The standard stock reagents (Merck, Germany) containing 1000 mg·kg^{-1} of metals were used. Calibration reagents containing between 0.5 and 5000 mg·kg^{-1} of Cd, Cu, Pb, and Zn in 3% HNO_3 were daily prepared.

2.3. Acid Leaching Experiment. In this study, five acid reagents were taken into account for leaching heavy metals from contaminated soil, including 0.05 M HNO_3, HCl, H_3PO_4, and H_2SO_4. A liquid-to-soil ratio of 1:10 was adopted by adding 5 g of soil with 50 mL above acid reagents in a glass vessel, and they were continuously stirred at 100 rpm for 5 h. After leaching experiments, the suspension was centrifuged at 3500 rpm for 30 min and then was filtered through a 0.2 μm membrane filter for metals analysis. The contents of metals were analyzed by ICP/AES. To reduce the systematic errors, duplicate measurements were carried out for each sample, and the average values were determined. A five-step sequential extraction procedure used in this study was developed by Tessier [20]. The five fractions of bound heavy metals included the following: exchangeable form (EXC) was extracted with 1 M NH_4OAc at pH 7, carbonates form (CAR) was extracted with 1 M NaOAc at pH 5, Fe–Mn oxide form (MNO) was extracted with 0.04 M $NH_2OH{\cdot}HCl$ in 25% HOAc, organic matter and metals associated with easily oxidizable solids or compounds (OM) were extracted with H_2O_2 (30%) at pH 2 and 0.02 M HNO_3, and residual form (RES) was extracted with a 5 : 1 mixture of HF-$HClO_4$. All extracted reagents were centrifuged for 30 min at 10000 rpm and filtered with a 0.2 μm Teflon filter. The suspensions were analyzed by ICP/AES. Soil particle size is an important factor to affect removal efficiency of heavy metals. However, it is difficult to screen soil particle size when the in situ remediation is throughout used to a real heavy metal-contaminated site. Therefore, the effect of soil particle size is not mentioned in this study.

2.4. Seed Germination Experiment. For the germination tests, seeds of cabbage (*Brassica chinensis* L.) were used because this species is a general vegetable and can be popularly seen for a whole year. Twenty-five seeds of cabbage were sowed in a sterilized individual tray ($15 \times 20 \times 3$ cm^3) containing 1200 cm [3] of modified leached soils. Duplicate of the seed germination experiment was simultaneously conducted. The trays were placed in a greenhouse at 25–28°C and 70% humidity. The experiment was examined daily for 30 days, and seeds were recognized to have germinated when the cotyledon was observed. After cotyledon appearance, the root lengths in each tray were measured and recorded every 12 h. The untreated soil sample was also conducted under the identical condition for comparison. To assess the seed germination criteria, the percentage of relative seed germination was chosen to understand the seed germination condition [21].

3. Results and Discussion

The physical and chemical properties of the soil samples along with their total heavy metal concentrations are listed

Table 1: The brief description of contaminated soil used in this study.

Property	Original soil
Texture	Sandy clay loam
pH	7.6 ± 0.11
Organic matter (%)	1.91 ± 0.18
CEC (cmol·kg^{-1})	4.58 ± 0.21
Total N (%)	0.13 ± 0.01
Total P (%)	2.82 ± 0.10
Total K (%)	1.74 ± 0.04
Available N (mg·kg^{-1})	22.4 ± 1.17
Available P (mg·kg^{-1})	187 ± 8.60
Available K (mg·kg^{-1})	764 ± 40.54
Total metal concentrations (average content, mg·kg^{-1})	
Zn	1334 ± 83
Cd	25 ± 3
Cu	263 ± 18
Pb	525 ± 31

in Table 1. The average total concentrations of Zn, Cd, Cu, and Pb were 1334, 25, 263, and 525 mg·kg^{-1}, respectively, and their maximum concentrations of Zn, Cd, Cu, and Pb were 4852, 46, 1470, and 1430 mg·kg^{-1}, respectively. The total heavy metal concentrations were not only higher than the tolerable limits recommended by the World Health Organization (WHO) and European Union (EU), but also exceeded the Chinese standard for agricultural soil [22]. This contaminated site needs to be appropriately treated to avoid heavy metals moving into deeper layers and causing phreatic zone pollution. As shown in Table 1, the organic matter content is much lower than the normal content. This is because that the Er-Jen River is contaminated by metal reclamation, smelting, and electrical capacitors and results in a high content of heavy metal accompanying a low pH value. Due to long-term irrigation from Er-Jen River, it is believed that the organic content of soil in this site is much lower than that of the uncontaminated site. The removal efficiency of heavy metals by various acid reagents is shown in Figure 1. The leaching experiments indicated that Zn and Cu had the highest removal efficiency and more than 70% removal efficiency was achieved for all acid reagents. For the Cd species, regardless of the acid reagents used, the removal efficiency ranged from 55% to 66%, indicating a high chemical affinity between the Cd and the soil, yielding lower removal efficiency for all acid reagents. A sequential extraction method was used to determine the Cd distribution in the surface sediment and found the major binding structure between Cd and soil was attributed to a residual form [23]. Among the four acid reagents, HNO_3 had the lowest Cd, Zn, and Cu removal efficiency, whereas H_2SO_4 had the highest Cd, Zn, and Cu removal efficiency; this result was not observed for Pb. To further understand the speciation distribution of heavy metal in soil, the chemical sequential extraction was used and is shown in Figure 2. The highest Cd concentration was associated with the RES fraction, whereas the EXC had the lowest concentration. The presence of Cd in the RES fraction was consistent with the result obtained by Mendoza et al. in which their report

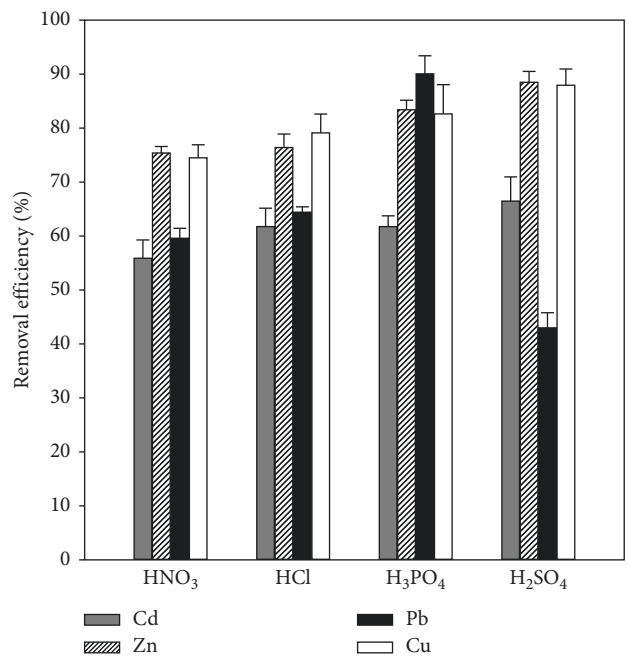

Figure 1: Removal efficiency of heavy metals by using various acid reagents.

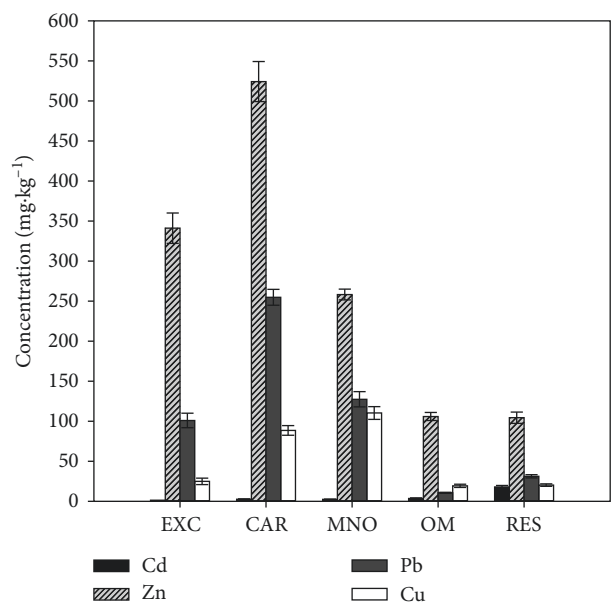

Figure 2: The sequential fractionations of heavy metals in contaminated soil.

showed that Cd was distributed predominantly in the residual form with a value of 45% in the contaminated soil [24]. Thus, the Cd retained in the crystal lattices of minerals has a strong bond and consequently is not easily released into the environment. The overall percentage of Cd in the fractions was in the order: RES > OM > CAR > MNO > EXC. Unlike Cd, the largest portion of Zn was observed in the CAR fraction (39%) and followed in the descending order of EXC > MNO > OM > RES. This fraction could be easily

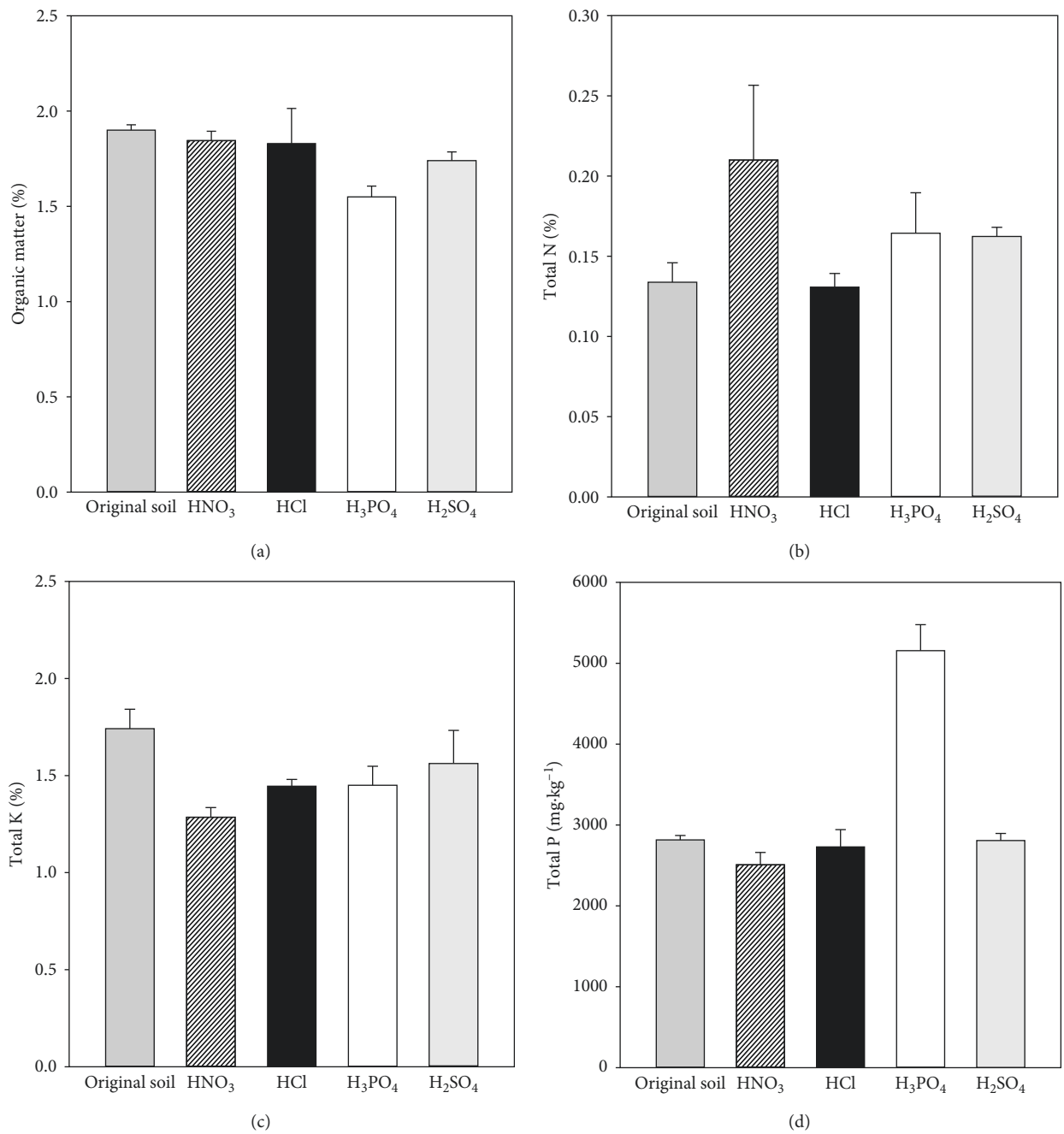

Figure 3: The changes in content of (a) organic matter, (b) total N, (c) total K, and (d) total P after leaching by various acid reagents.

made exchangeable under conditions such as pH change [25]. Pb and Cu demonstrated an identical trend in that CAR and MNO appeared to be the major fraction. The proportion of heavy metals bounds to Fe-Mn oxides was highly variable and depended on the depth of the water and redox reactions with the soils. The redox cycle is important in controlling Fe and Mn in most aquatic and soil systems. However, it varies seasonally in summer, when the oxygen between the water and soil interface often decreases. This causes the reduction of Fe^{3+} and Mn^{4+} into soluble Fe^{2+} and Mn^{2+}, which is transported upward in the water column and oxygenated, resulting in the reoxidation of insoluble metals to settle the soil and repeat the cycle [26]. These forms of metals are potentially dangerous for plants because they can transform into being bioavailable under environmental changes [27]. Organic matter (OM), nitrogen (N), potassium (K), and phosphor (P) are important substances for vegetable and plant growth in soil. To understand the changes in these substances after acid reagents leaching, OM, total N, total K, and total P contents were determined and are depicted in Figure 3. The content of OM in the soil decreases by 18.5% and 8.74% when H_3PO_4 and H_2SO_4 were used for pickling, respectively. No significant change in the OM content for HNO_3 and HCl was observed. The total N content increased

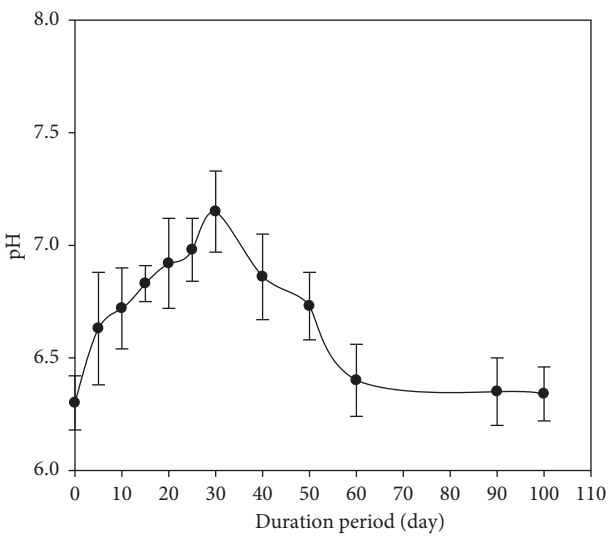

Figure 4: The change in soil pH as a function of the duration period after HCl leaching and fertilized by adding organic manure.

by 57.07%, 22.86%, and 21.47% after leaching with HNO_3, H_3PO_4, and H_2SO_4, respectively. The N content decreased by 10.65% after leaching with HCl. Higher N content after HNO_3 treatment may be attributed to the presence of NO_3^- released from HNO_3 during the leaching process. Figure 3(d) shows the phosphorus content in the soil after acid leaching. All reagents, except H_3PO_4, reduced the total P content after acid leaching. No significant difference was noted between the original soil and the other three acid reagents. The P content increased by 83.17% after leaching by H_3PO_4, presumably because of the presence of PO_4^{3-} released from the H_3PO_4 during leaching process. The result of changes in total K is also depicted in Figure 3(c). The soil K concentration changed because of acid liquid precipitation. HNO_3 reduced the total K content by 26.2%, followed by others in a descending order $HCl > H_3PO_4 > H_2SO_4$. Commercial organic manure was used after the leaching treatment to fertilize the soil. Commercial organic manure contains 21% OM, 4.2% total N, 4.1% total P, and 3.5% total K. Most manure containing considerable amounts of soluble salts increases soil's electrical conductivity (EC), resulting in damage to plants. The result of the experiments showed that the EC increased after manure treatment and reached stability after 30 days. Over 30 days, the EC ranged from 0.86 to 0.41 $mS \cdot cm^{-1}$, an acceptable range for plant growth. Figure 4 shows the changes in soil pH as a function of duration after HCl leaching and fertilization by organic manure. The initial pH for original and leached soils was 7.6 and 6.3, respectively. The acid leaching treatment reduced the soil pH consequentially caused an unfavorable effect to crops cultivation. The pH increased with the duration and reached a maximum of 7.02 at 30 days; it then gradually decreased and eventually stabilized to approximately 6.3. The addition of organic manure significantly improved soil pH over a short period; however, in the long-term, the soil pH returned to a meta-acid state, which was probably associated with the soil buffering power. Figure 5 shows the changes in the OM, total N, total P, and total K contents as a function of the duration after HCl leaching and fertilization treatment. The identical feature could be observed after the addition of organic manure, which increased the OM, total N, total P, and total K content within 30 days. Over the first 15 days, the OM and total N contents became more than twice of this manure addition, and a steady state was gradually achieved. However, the total K content rapidly increased with the duration with the highest content value of 2.5% within 5 days. On the basis of these findings, the organic manure additive improves soil fertility within a short period. To assess the suitability of farmland for cultivation, a seed germination experiment was performed to evaluate suitability for planting after various treatments. Seeds were cultivated in an incubator and observed daily for germination with the seed germination rate as a function of time at various treatments presented in Figure 6. The lowest germination rate was observed when soil was treated through acid leaching because after acid leaching the soil fertility decreased and affected plant growth. On the other hand, initial seed germination occurred earlier than the other treatments. Although heavy metals were moderately removed after acid leaching, the nutrient loss was a major barrier to seed germination. After the addition of organic manure, the seed germination rate considerably increased by more than 80% and the initial seed germination time was four days, earlier than that of the experiments assessed after acid leaching. Notably, the seed germination rate in untreated soil was superior to that in acid-leached soil. This demonstrates that the inhibition effect of acid leaching to the plant is more serious than that of heavy metals, and the addition of organic manure could be a suitable approach to improve soil fertility and plant cultivation. To obtain a higher seed germination rate, the procedure of soil turnover was applied simultaneously. As shown in Figure 6, the procedure of adding manure to soil turnover rapidly increased the seed germination rate and initial seed germination time, and a seed germination rate of approximately 85% was achieved. The procedure of soil turnover diluted heavy metals and acidic species and thus resulted in the redistribution of these species.

Although seed germination rate is a suitable indicator to evaluate soil fertility, heavy metal concentration in test plants should be considered to ensure food safety. Therefore, cabbage roots and leaves were collected and analyzed to understand heavy metal distribution. The experimental result is presented in Table 2. The major uptake was found in the root of cabbage, and the fraction of Cd, Zn, Pb, and Cu uptake by the root was 46.8%, 51.3%, 17%, and 25.4%, respectively. This finding is in agreement with the observation of Shi et al., who reported that heavy metal concentration is much higher in the root than in the shoot and leaf [28]. The root is the first mode of entry for heavy metal pollutants from the soil into the plant. Nevertheless, it excretes organic acids, amino acids, sugars, and growth substances into the rhizosphere, affecting metal adsorption. By changing the physiochemical properties of the rhizosphere, the heavy metals will be detained in the roots through chelation, complexation, and disposition with root exudates. Notably,

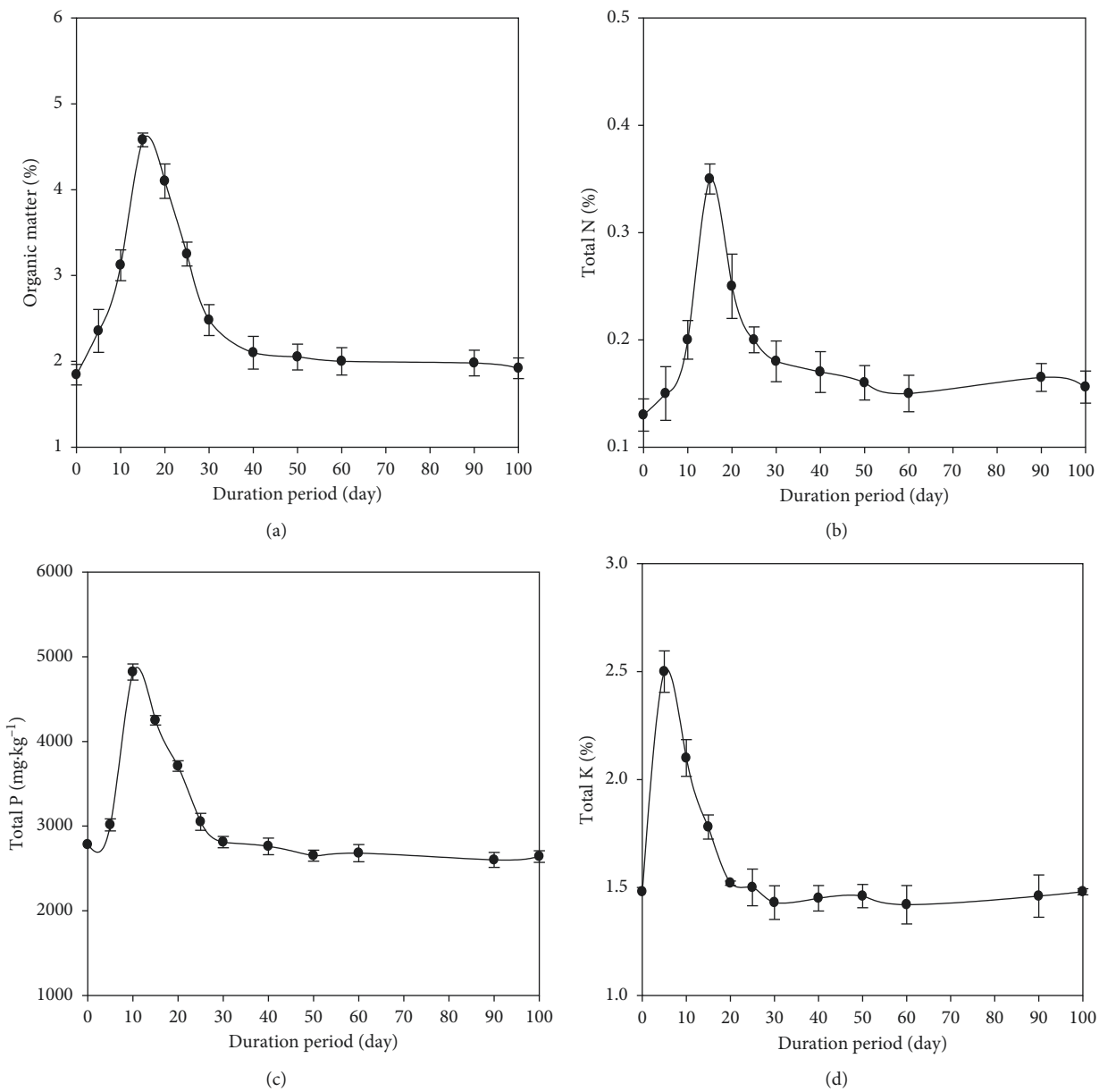

Figure 5: The changes in contents of (a) organic matter, (b) total N, (c) total P, and (d) total K as a function of the duration period after HCl leaching and fertilized by organic manure.

in this case, the Pb uptake by the root was the lowest, whereas a much higher fraction was noted for Zn; the leaf demonstrated the same result. Pb and Zn are essential elements for plants growth, but their excessive amounts in soil lead to the inhibition and poisoning of plants. The normal Pb, Cu, and Zn contents in plants are 0.5–10 mg·kg^{-1}, 3–30 mg·kg^{-1}, and 10–150 mg·kg^{-1}, respectively, whereas their toxic concentrations to plants are 100–400 mg·kg^{-1}, 70–400 mg·kg^{-1}, and 60–125 mg·kg^{-1}, respectively [29]. Here, the Pb, Zn, and Cu concentrations in bulk cabbage were 120.23 mg·kg^{-1}, 283.52 mg·kg^{-1}, and 37.14 mg·kg^{-1}, respectively. Except Cu, Pb and Zn concentrations were slightly higher than the aforementioned range, indicating the soils after acid leaching and organic manure addition remain a risk when used to cultivate plants. The procedure of soil turnover may be a suitable approach to overcome this obstacle.

4. Conclusions

A series of acid leaching tests and soil fertility assessment for contaminated soil containing Cd, Pb, Zn, and Cu were performed to investigate leaching behavior. Most heavy metals could be removed through acid leaching with a nearly 60% removal efficiency achieved. The major species for Cd, Zn, and Cu were distributed in residual, bound-to-carbonate, and bound-to-Fe-Mn oxides forms, respectively. The addition of organic manure distinctly increased soil nutrient content as well as soil pH within a short period. Seed

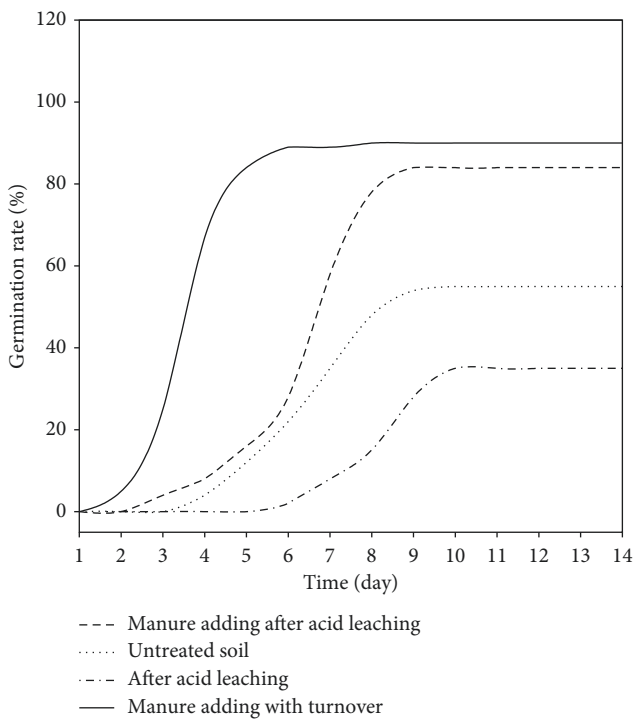

Figure 6: Seed germination rate as a function of time at various treatments.

Table 2: Distribution of heavy metal concentration at the root and leaf of cabbage.

	Cd (mg·kg^{-1})	Zn (mg·kg^{-1})	Pb (mg·kg^{-1})	Cu (mg·kg^{-1})
Root	1.47 ± 0.04	145.52 ± 9.84	20.45 ± 1.74	9.42 ± 0.87
Leaf	0.13 ± 0.03	65.85 ± 2.42	4.33 ± 0.13	3.84 ± 0.27
Bulk	3.14 ± 0.21	283.52 ± 12	120.23 ± 8.87	37.14 ± 3.14

germination in untreated soil was superior to that in acid-leached soil, revealing that the phytotoxic effect of acid leaching is more serious than that of heavy metals.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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