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Remediation of Heavy Metal Contaminated Farmland Soil by Biodegradable Chelating Agent GLDA

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Abstract: Chemical leaching is one of the effective methods to remove heavy metals from soil. The effects of biodegradable chelating GLDA (*N*, *N*-bis(carboxymethyl)-L-glutamic acid tetrasodium salt) on contaminated farmland soil in four different places (collected from Shangba Village, Shaoguan city (SB); Huaqiao Village, Zhuzhou (HQ); Shaoguan Tielong (TL); and Liantang Village, Lechang (LT), respectively) were studied by the method of leaching. To explore the synergistic effect between GLDA and citric acid, the leaching conditions were also explored. The results showed that the leaching efficiency of heavy metal Pb in soil was improved by adjusting the GLDA solution to reach acidity by adding citric acid. The leaching efficiency of Pb after mixing GLDA and citric acid was higher than the sum of their respective leaching abilities. After leaching with 10 mmol/L of the GLDA solution with a pH of 10.86 for 120 min, the total Pb and the Pb available in the soil decreased significantly. It is known that citric acid has a significant effect on improving the leaching rate of GLDA on soil, and this method can also be applied to other heavy metals, such as Cd. This study provides a low-ecological-risk method for the remediation of heavy metal-contaminated soils.

Keywords: Pb; soil leaching; GLDA; citric acid; heavy metals

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1. Introduction

In recent years, heavy metal soil pollution caused by various activities, including agricultural practices, industrial activities, and waste treatment, has become a problem of worldwide concern [1–3]. Environmental soil problems are becoming more and more serious in China [4,5]. In the remediation of heavy metal-contaminated soil, leaching with synthetic extracts, such as aminopolycarboxylic acid chelators (aminopolycarboxylate chelants, APCs), is one effective method of removing heavy metals from the soil. It uses physical processes or both physical and chemical processes to limit pollutants in the soil [6,7], and it is able to interact with most toxic metals. Common chelating agents include EDTA, EDDS, GLDA, NTA, and HIDS—among which, EDTA is the environmentally durable chelating agent, and the other three are biodegradable chelating agents [8]. EDTA is considered to be the most effective chelator for heavy metal removal [9–11]. This is because it can form strong complexes with divalent and trivalent metals and alkaline earth metals, such as the macronutrients calcium (Ca) and magnesium (Mg), and trace elements, such as iron (Fe), copper (Cu), zinc (Zn), nickel (Ni), lead (Pb), and cadmium (Cd) [12,13]. It is often used as a preservative or stabilizer in cosmetics and as a food additive. However, EDTA is not easy for the environment to biodegrade, and it persists for a long time. Moreover, it is highly persistent in aquatic environments and has potential ecological risks [14]. High concentrations of EDTA may be toxic to soil, aquatic organisms, and plants [14]. It can improve bioavailability and phytotoxicity in sewage sludge or contaminated soil by changing the permeability of cell membranes [15]. Therefore, scholars and others are also constantly studying biodegradable chelating agents, and more and more people are paying attention to the development and use of environmentally friendly chelating agents with better biodegradability and less environmental toxicity [16–18]. At present, EDDS has become the main alternative product, which degrades easily [18,19]; however, EDDS is expensive, and the degradation effect is not as good as EDTA [20]. The cost of HIDS is high, and NTA has adverse effects on health [1,21]. At the same time, the latest research shows that nano-sized adsorbents, such as BFR and SBR, have also been applied in the adsorption treatment of wastewater and soil, which can effectively remove toxic metals from the environment [5].

A new biodegradable, environmentally friendly chelating agent, GLDA (*N*,*N*-bis(carboxymethyl)-L-glutamic acid tetrasodium salt), has attracted attention [22,23]. It is considered to have the greatest potential to purify soil by in situ washing treatment compared to other biodegradable chelators, and its process is based on green chemistry [24]. The traditional biodegradability test (OECD 301D) shows that its consistent degradability far exceeds the 60% degradation rate that is considered to be easily biodegradable. Biodegradable chelating agents, HIDS and GLDA, can better solve the problem of ecological risks to the environment during the use of EDTA and are often used to recover potentially toxic elements or rare metals from various solid wastes [6]. GLDA is mainly made from plant materials [25]. Different from the production of EDTA, the carbon source of GLDA is mainly biological. Compared with other chelating agents, GLDA has the smallest ecological footprint (a smaller ecological footprint means a smaller impact on the environment) and is easy to biodegrade. In terms of chelating capacity, GLDA is as efficient as EDTA [6] when used to remove heavy metals from soil and sludge [7,26,27]. GLDA has good chelation ability for many metal ions and good biodegradability. More than 60% of GLDA can be degraded within 28 days [28]. During soil leaching and remediation, GLDA is often combined with other chelating agents to form a mixed chelating agent [29,30]. However, up until now, studies have mainly focused on EDTA, and there are few studies on the biodegradable chelator, GLDA. There are few studies on the pH regulation of GLDA by citric acid, the influence of different leaching times of GLDA, and the amount of GLDA on the leaching effect in the relevant literature.

During the study, various parameters, such as the contact time, pH value, chelator concentration, stability constant, and metal species distribution, affected the metal performance expression of chelators. In the experiment, we used the complexation reaction of heavy metals with biodegradable chelating agents to remove heavy metals from the soil and tested the physical and chemical properties of the soil before and after removal. In order to further improve the leaching removal efficiency of Pb from the soil by GLDA, and to explore the influence of different pH levels on the leaching effect of heavy metals in the soil by GLDA solution, citric acid and a GLDA solution were mixed, and four soil samples from different places were selected for soil leaching. The leaching effect of the combination of GLDA and citric acid was explored to determine whether it was better than that of the separate leaching effects. Further, on this basis, the influence of dosage and oscillation time on the leaching effect was explored, as well as the effect of the combination of GLDA and different salts, so as to determine the leaching condition with the best effect and the lowest cost. In this study, the effect of GLDA on the removal of heavy metal Pb from the soil by soil leaching under different application conditions was discussed for the first time. This method can be applied not only to metal Pb but also to other metals, such as Cd.

2. Experimental

2.1. Materials

Soil Sample

Surface soil samples were collected from the polluted farmland in Shaoguan Shangba (SB), Zhuzhou Huaqiao (HQ), Shaoguan Tielong (TL), Lechang Liantang (LT), and other places, and the basic physical and chemical properties were shown in Table 1. The soil samples were dried naturally in the laboratory, and the stones and other sundries in the samples were removed. After grinding, the soil samples were stored in a clean sealing bag through a 20-mesh or 100-mesh nylon sieve, according to different requirements.

Table 1. Basic physical and chemical properties of the soil sample.

	pH	**CEC**	**OM**	**Fe**	**Pb (S = 406.9)**	**Cd**	**Cu**	**Zn**
		cmol/kg	**g/kg**	**μg/g**	**μg/g**	**μg/g**	**μg/g**	**μg/g**
Dabaoshan SB	5.04	8.87	13.26	50855.02	287.26	0.58	210.02	249.83
Huaqiao HQ	5.01	10.07	41.06	12364.12	42.87	0.71	25.53	71.41
Tielong TL	6.01	8.54	30.32	18826.91	146.08	3.30	27.49	198.72
Liantang LT	7.17	13.63	29.63	38993.00	947.63	1.87	43.56	954.50

2.2. Reagent

The biodegradable chelating agent, GLDA (*N*, *N*-bis(carboxymethyl)-L-glutamic acid tetrasodium salt, GLDA-Na4, 47% aqueous solution, product name-Dissolvine GL-47, purchased from Linsilen, hereinafter referred to as GLDA-Na) [31], citric acid, sodium hexametaphosphate, sodium citrate, ferric chloride, and potassium chloride are analytically pure.

2.3. Experimental Procedures

Heavy metal extraction experiments were carried out at a solid-to-solution ratio of 1:20. For each run, a soil sample (1.00 g) was suspended in a 20 mL solution and was agitated using an end-over-end shaker at a speed of 200 rpm at room temperature for a given time. The suspensions were centrifuged at 3500 rpm for 10 min and filtered through a 0.45 μm membrane filter. The concentrations of Pb were measured by AAS (Hitachi Z-2300). All tests were performed in triplicates.

The validation of the developed methods was carried out through the analysis of the parameters of linearity, detection limits (LOD), quantification limits (LOQ), accuracy, and precision, using sophisticated statistical tests in accordance with recommendations in the literature [32,33].

2.4. Distribution of Toxic Metals in the Contaminated Soil before and after Chelant Extraction

A selective sequential extraction (SSE) procedure has been carried out to determine the mobility and dissemination of toxic metals in the organic-rich contaminated soil sample before and after treatment with the chelants. The employed SSE scheme was developed by the Standard Measurements and Testing Programme (formerly BCR) of the European Community, and the metal distribution in the contaminated soil was operationally defined as—(a) exchangeable, water- and acid-soluble, (b) reducible, (c) oxidizable, and (d) residual fractions. The working protocol of the SSE scheme is described elsewhere [34,35].

2.5. Leaching of Soil with GLDA Solution of Different pH

The pH of 5 mmol/L of GLDA-Na solution was adjusted to 4, 5, 6, 7, and 10 by 0.4 mol/L citric acid solution, denoted as GLDA-4, GLDA-5, GLDA-6, GLDA-7, GLDA-10, and GLDA solution without adjusting pH (pH 11.13). Distilled water was added with the same amount of citric acid (CA) for regulation, denoted as CA-4, CA-5, CA-6, CA-7, and CA-10, respectively.

One gram of the contaminated soil samples from four different places (SB, HQ, TL, and LT) was weighed and loaded into 100 mL centrifuge tubes. Then, 20 mL of the above 11 solutions (GLDA-4, GLDA-5, GLDA-6, GLDA-7, GLDA-10, GLDA, CA-4, CA-5, CA-6, CA-7, and CA-10) were added and shaken at room temperature for 2 h (200 r/min).

2.6. Leaching of Soil with Different Leaching Times and Different Dosages of GLDA

GLDA-4 and GLDA-Na were selected as leaching solutions with a concentration of 5 mmol/L, and leaching was carried out at four oscillation times of 30 min, 60 min, 120 min, and 240 min. The rest was the same as above.

The concentrations of the GLDA-4 and GLDA-Na solutions were set to 2.5, 5, 10, and 20 mmol/L as the leachate, shaken for 120 min, and the rest was the same as above.

The removal rate of heavy metals was calculated according to the equation:

Removal rate of heavy metals (%) = removal amount of heavy metals/total amount of heavy metals in the soil × 100%.

2.7. Effects of Combinations of GLDA and Different Salt on the Removal of Heavy Metals in Soil

Different salts were selected and combined with GLDA-Na, as shown in Table 2. The concentration of the GLDA solution was 5 mmol/L, and the sodium hexametaphosphate, sodium citrate, and potassium chloride solution was 0.1 mol/L. Then, 10 mmol/L of GLDA-Na was mixed with 0.2 mol/L salt solution at a volume of 1:1.

Table 2. Combination of GLDA and different salts.

Dispose	Concentration
GLDA	5 mmol/L
Sodium hexametaphosphate	0.1 mol/L
G-Sodium hexametaphosphate	10 mmol/L GLDA-Na was mixed with 0.2 mol/L sodium hexametaphosphate in equal volume
Sodium citrate	0.1 mol/L
G-Sodium citrate	10 mmol/L GLDA-Na was mixed with 0.2 mol/L sodium citrate in equal volume
Potassium chloride	0.1 mol/L
G-Potassium chloride	10 mmol/L GLDA-Na was mixed with 0.2 mol/L potassium chloride

2.8. Leaching of Contaminated Soil by GLDA-4

Soil samples of SB, TL, and LT were selected. Then, 400 mL of eluent GLDA-4, 10 mmol/L (pH = 10.86) was added to the 20 g soil sample and shaken (200 r/min) for 120 min. After the soil was washed with GLDA-4, the washed soil was washed with distilled water (the same amount as the eluent) 3 times. The soil samples were air dried and used for backup, and the pH of the soil was measured.

2.9. Data Processing

Excel 2016 was used for data analysis, processing, and plotting, Duncan's analysis method in SPSS Statistics 22 was used for statistical analysis, and the significance level for evaluating the difference between treatments was set as $p < 0.05$.

3. Results and Discussion

3.1. Leaching Effects of GLDA and Citric Acid Solutions with Different pHs on Soil Heavy Metals

The chemical structure and basic information of GLDA-Na are shown in Figure 1. The pH of the GLDA-Na solution was adjusted with the citric acid solution. Under different pH conditions, the leaching effect of GLDA-Na on Pb in the soil is shown in Table 3. Compared with the GLDA-Na solution without adjusting pH, the leaching rate of Pb in soil with the GLDA-Na solution at pH levels of 4 and 5 was significantly increased. After adding citric acid to adjust the GLDA-Na solution to reach acidity, the leaching efficiency of Pb from soil was improved [33]. The leaching effect of GLDA-Na and citric acid on Pb was better than the sum of the GLDA-Na and citric acid effects (Table 3).

For the contaminated soil in Lechang, the leaching rate of Pb by GLDA-4 was 48.45%, which was greater than the sum of the leaching rate of GLDA-Na without pH adjustment (11.40%) and C-4 (8.13%). The leaching rate of GLDA-5 (40.86%) was greater than that of GLDA-Na (11.40%) + CA-5 (4.60%). The leaching rate of GLDA-6 (33.12%) > GLDA-Na (11.4%) + CA-6 (0.48%), GLDA-7 (27.71%) > GLDA-Na (11.40%) + CA-7 (0.54%) without pH regulation. Finally, the leaching rate of GLDA-10 (21.67%) > GLDA-Na (11.40%) + CA-10 (0.02%). The soil performance trends in Lechang, Tielong, Zhuzhou Huaqiao, and Dabaoshan were consistent.

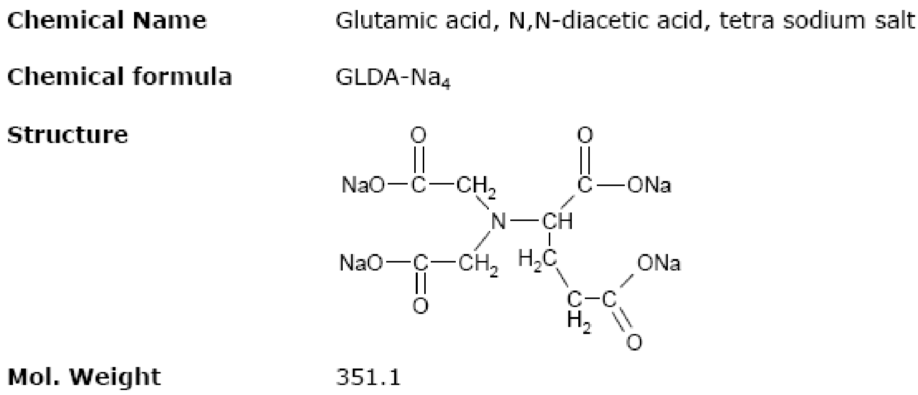

Figure 1. The structure of GLDA.

Table 3. Leaching removal rate of heavy metal Pb under different treatments (%).

Leacheates	SB	HQ	TL	LT
GLDA-4	38.74 ab	51.65 a	56.70 a	48.45 a
GLDA-5	39.91 a	49.68 a	48.29 b	40.06 b
GLDA-6	37.79 b	46.84 b	45.20 c	33.12 c
GLDA-7	38.93 ab	42.48 c	42.78 d	27.71 d
GLDA-10	35.73 c	43.46 c	38.89 e	21.67 e
GLDA	23.98 d	33.65 d	30.09 f	11.40 f
CA-4	6.96 e	7.65 e	5.76 g	8.13 g
CA-5	4.47 f	4.80 f	3.44 h	4.60 h
CA-6	2.07 g	2.11 g	1.81 hi	0.48 i
CA-7	1.95 g	2.10 g	1.65 hi	0.54 i
CA-10	0.11 h	4.40 g	0.70 i	0.02 i

According to Duncan's multiple range test ($p < 0.05$), the means in the same line (same contaminated soil) followed by the same letter were not significantly different.

3.2. Influence of Leaching Time on the Leaching Effect of Heavy Metal Pb by GLDA-Na

When the soil was washed with a GLDA-Na solution with a pH of 4 and an unadjusted pH, the removal rate of heavy metals increased over time, and after 120 min, the removal rate of heavy metals tended to be stable (Figures 2 and 3). Therefore, the best leaching time could be selected as 120 min. When the pH was 4, the complexation effect of the GLDA with heavy metals was better and the removal rate was high at 120 min. However, the reduction rate decreased after the time exceeded 120 min; therefore, it was inferred that the complexation of heavy metals with chelating agents basically reached equilibrium and reached the maximum value during 120 min, and the extraction efficiency could not be improved by prolonging the time. The leaching efficiency of the GLDA-Na solution at a pH of 4 was higher than that without adjusting the pH. At 120 min, the leaching efficiency of GLDA-4 on the Huaqiao soil was 51.65%, and that of the GLDA-Na solution without adjusting the pH was 33.65%. At 30 min, the leaching removal rate of the GLDA-4 was 37.15%, which exceeded the highest leaching efficiency of GLDA-Na without adjusting the pH value.

The soil in the other three sites showed the same trend. It could be seen that to achieve the same efficiency, the leaching time of GLDA-Na with a pH value of 4 is greatly shortened. The stability of metal-chelating agent complexes depends on many factors, including the oxidation state and coordination number of metal ions, as well as the electronic structure and properties of the chelating agents. However, these factors no longer change significantly after a certain period of contact [6].

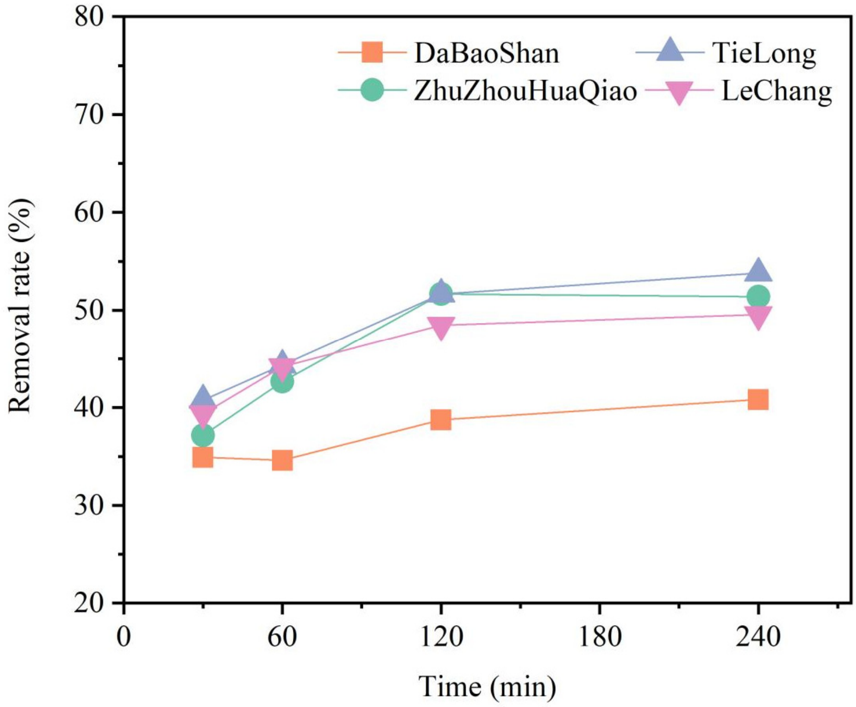

Figure 2. The change of the Pb leaching removal rate with the time of GLDA-Na with pH 4 and 5 mol/L concentration.

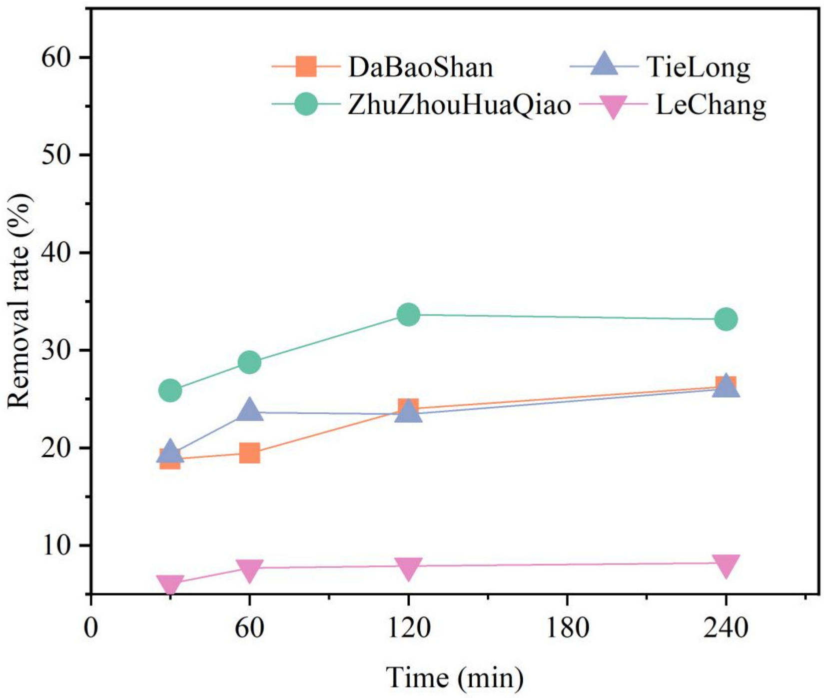

Figure 3. The change in Pb leaching removal rate with time of GLDA-Na with pH 11.13 of 5 mol/L concentration.

3.3. Effects of GLDA-Na Solutions with Different Concentrations on the Removal of Heavy Metal Pb in Soil

With the increase in the concentration of the GLDA-4 leaching solution, the removal of heavy metal Pb was first increased rapidly, and then the variation was small (Figure 4). For the same concentration of the leaching solution, the leaching efficiency at pH 4 was higher than that of the unregulated GLDA solution (Figures 4 and 5). At 10 mmol/L, the leaching rate of Pb in the Huaqiao soil was 48.48% with GLDA-4 and 33.69% with GLDA without adjusting the pH. At 5 mmol/L, the leaching removal rate of GLDA-4 was 40.57%, which exceeded the highest removal rate of the GLDA solution without adjusting the pH. Hence, in order to achieve the same efficiency, the amount of leaching of the GLDA solution with a pH of 4 was greatly reduced. When the concentration increased, the removal rate decreased rather than increased, which we speculated to be related to the soil pH and the total amount of heavy metals in the soil. When the number of chelating agents added was lower than the amount needed by the soil, the reduction rate would increase, but when too many chelating agents were added, the reduction rate would decrease [22].

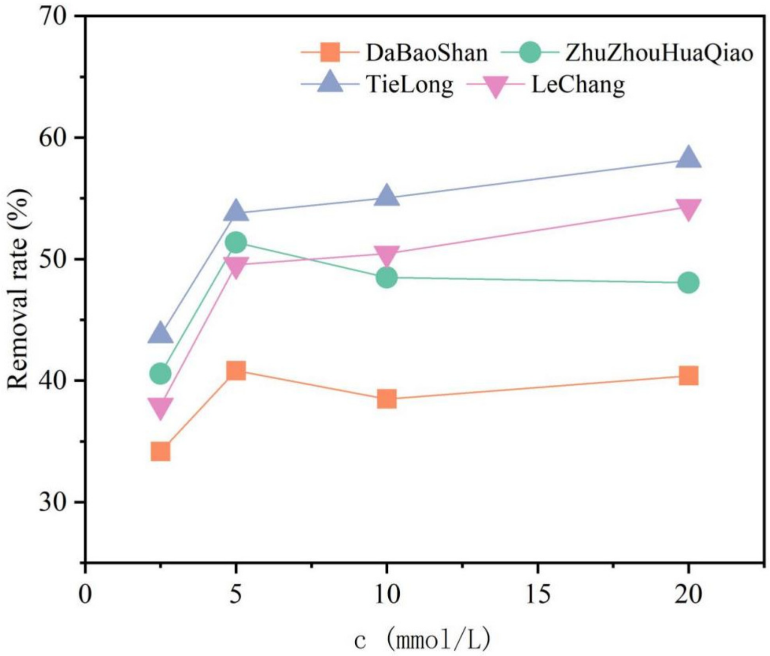

Figure 4. The leaching removal rate of Pb varies with the concentration of GLDA-Na with a pH of 4 after 120 min.

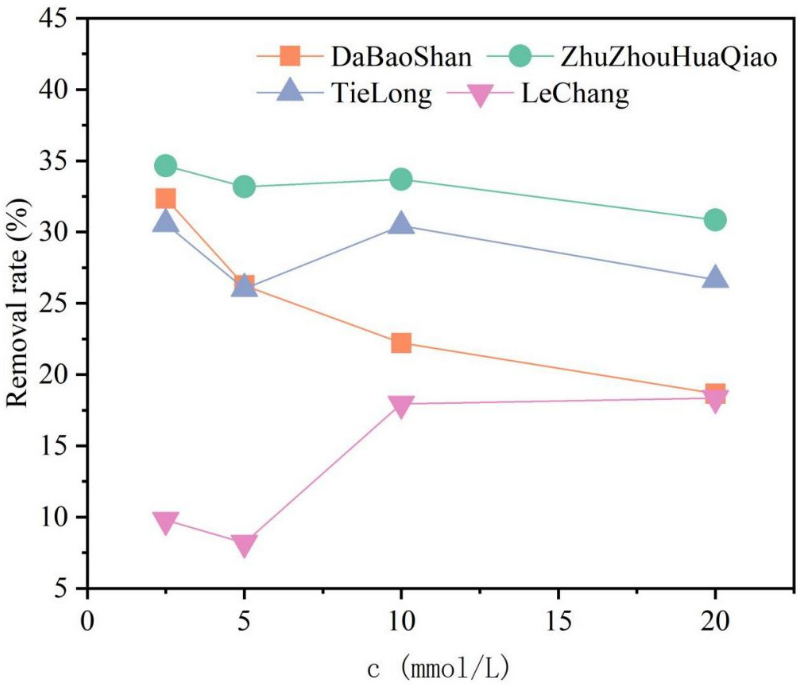

Figure 5. The leaching removal rate of Pb varies with the concentration of GLDA-Na with a pH of 11.13 after 120 min.

3.4. The Effects of Combinations of GLDA-Na and Different Salts on the Removal of Heavy Metals in Soil

Separate leaching of different salts could remove Pb in soil (Table 4), and the removal rates from high to low were sodium hexametaphosphate, sodium citrate, and potassium chloride. Adding sodium hexametaphosphate into the GLDA-Na solution could improve the leaching rate of Pb in soil, but it did not produce the effect that 1 + 1 was greater than 2. Sodium citrate and potassium chloride could not improve the leaching efficiency of GLDA-Na. It could be seen that the combination of sodium hexametaphosphate, sodium citrate, and potassium chloride with GLDA-Na could not significantly improve the leaching efficiency of GLDA-Na.

Table 4. Leaching removal rate of Pb in the soil after the combination of GLDA-Na and different salts (%).

	GLDA-Na	Sodium Hexametaphosphate	G-Sodium Hexametaphosphate	Sodium Citrate	G-Sodium Citrate	Potassium Chloride	G-Potassium Chloride
Dabaoshan	23.98	29.61	30.83	3.73	15.73	1.98	20.53
Huaqiao	33.65	40.39	44.58	10.47	28.67	2.63	34.54
Tielong	30.09	39.49	41.11	4.53	22.13	0.00	26.96
Lechang	11.40	34.09	35.70	0.71	6.70	0.00	6.81

3.5. Soil Condition after Leaching with GLDA-4

GLDA-Na solution with a pH of 4 was selected, leaching time was 120 min, leaching concentration was 10 mmol/L, and the soil was washed. The full amount of Pb, available content, and pH are shown in Table 5. After leaching, the total Pb content in the Dabaoshan, Tielong, and Lechang soils decreased by 44.0%, 74.9%, and 46.2%, respectively, and the available Pb contents decreased by 54.8%, 64.7%, and 69.9%, respectively. Leaching with GLDA-4 significantly reduced the total and available contents of Pb in the soil. The leaching of soil with GLDA-4 had little effect on the pH of soil from Dabaoshan and Tielong, and the pH of soil from Lechang decreased from 7.17 to 6.79.

Table 5. The heavy metal Pb content and pH of soil before and after leaching.

	Full Amount (mg/kg)		Available Content (mg/kg)		pH
Before leaching					
SB	287.3 ± 6.5		45.53 ± 4.34		5.04
TL	146.1 ± 17.6		42.877 ± 8.765		6.01
LT	947.6 ± 25.9		242.4 ± 10.4		7.17
After leaching		Reduction rate		Reduction rate	
SB	160.9 ± 8.4	43.97%	20.60 ± 0.75	54.73%	5.00
TL	36.7 ± 11.5	74.92%	15.11 ± 2.35	64.67%	6.02
LT	509.5 ± 11.3	46.23%	72.97 ± 6.29	69.89%	6.79

Solution pH is expected to affect the chelant capability of metal extraction by controlling the aqueous metal species concentration, solubility of the chelants, sorption or desorption and ion-exchange behavior of the metal ions, and re-adsorption mechanisms of the newly formed metal–chelant complexes [36,37]. The pH of the washing solution may affect the soil's retention of metals by adsorption and affect the capability of the chelator to remove metals by regulating the stability constants of the metal chelant complexes, the aqueous metal species concentration, metal sorption/desorption, and ion exchange [38,39].

4. Conclusions

(1) The leaching efficiency of heavy metal Pb in soil was improved by adjusting the GLDA-Na solution to reach acidity with citric acid.

(2) The leaching efficiency of Pb, achieved by mixing GLDA-Na and citric acid, was greater than the sum of their respective leaching abilities. To achieve the leaching

efficiency of GLDA-Na without adjusting the pH, the leaching time and the dosage of the GLDA-Na solution, adjusted by citric acid at a pH of 4, were reduced.

(3) After leaching with 10 mmol/L of GLDA-4 solution for 120 min, the total Pb and available Pb in the soil decreased significantly, with decreased amplitudes of more than 44% and more than 54%, respectively.

(4) The leaching rate of Pb was improved by adding sodium hexametaphosphate into GLDA-Na, but the effect of 1 + 1 > 2 could not be achieved.

(5) The combination of GLDA and critic acid is an environmentally friendly and effective chelating agent or washing solution for the remediation of heavy metal-contaminated soils. This mixture may be a useful, effective, and environmentally friendly chelator for the remediation of heavy metal-contaminated soil.

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