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Leaching Remediation of Dredged Marine Sediments Contaminated with Heavy Metals

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Abstract: There are more than 150 ports in China and a considerable proportion of dredged sediments in ports and waterways are contaminated with heavy metals as the typical contaminants. It is mandatory to remediate the contaminated dredged sediments prior to further resource utilization. The over-arching objective of this study was to use natural organic acids (oxalic acid, citric acid, tartaric acid, and malic acid) as leaching agents to remove heavy metals (Cu, Cd, and Pb) from contaminated dredged sediments. Batch experiments were conducted to investigate the factors governing the removal rate of heavy metals and leaching kinetics. Citric acid had the best leaching effect on heavy metals Cu, Cd, and Pb with an optimal leaching concentration of 20 mmol/L and a solid-to-liquid ratio of 1:20. The average removal rates of Cu, Cd, and Pb were 85%, 73%, 56%, and 35% for citric acid, malic acid, tartaric acid, and oxalic acid, respectively. The leaching kinetics showed that the removal of heavy metals increased rapidly with time and then gradually reached the maximum value which was best described by the Elovich equation model. The outcomes of this study suggest that citric acid is an effective and environmentally friendly leaching agent for removing heavy metals from marine dredged sediments.

Keywords: marine sediment pollution; environmental remediation; natural organic acid; batch experiments; ports and waterways

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1. Introduction

Ports and waterways are rarely natural deep-water areas. To meet the needs of shipping development, it is necessary to conduct dredging activities to continuously increase berth tonnage and ensure navigation safety. China is the world's largest country in terms of dredged volume in the ocean [1], with nearly two billion cubic meters of dredged volume in ports and waterways in 2019, and this is primarily disposed of through marine dumping and hydraulic reclamation. The continuous expansion of the dredging scale has led to a huge amount of dredged sediments, and the disposal of dredged sediments in China has become a prominent problem that not only creates issues for marine environmental protection but also increases the difficulty of marine management [2,3]. According to the level of contaminants, dredged sediments can be divided into three categories: clean dredged sediments, fouled dredged sediments, and contaminated dredged sediments [4]. Different types of dredged sediments have different degrees of impact on the surrounding marine environments. Clean dredged sediments have no or nearly no impact on the water quality of the overlying waters, and the dissolved contaminant concentration generally does not exceed the Marine Sediment Quality (GB 18668-2002, China). Therefore, clean dredged sediments can be directly dumped into the sea for disposal. Fouled or contaminated dredged sediments have a significant impact on the overlying seawater and benthic ecosystem, and this fact has attracted widespread attention in marine environments [5].

The Yingkou port is located on the northeast coast of Liaodong Bay in the Bohai Sea and is an important transportation hub port for coal, steel, ore, and crude oil in China. The early rapid expansion of the urban and industrial sectors of the Yingkou port area, including the booming petrochemical, steel, and coal transportation markets, discharged significant volumes of effluent into the coastal waters [6]. Steel, plastics, oils, ores, etc., are common sources of the exposure of heavy metals to the environment [7]. Consequently, a variety of contaminants had been reported in the dredged sediment samples of Yingkou Port, among which heavy metals (e.g., Cu, Cd, and Pb) are the most typical contaminants [8]. Heavy metal pollution in sediments/soils has become a hot issue due to the deficiency of the criteria between heavy metal concentrations and their ecological or human health impacts [9]. For biological processes, Cu is essential, but it can also have negative and toxic impacts at elevated levels [10]. By comparison, Pb and Cd are non-essential but highly poisonous, and their pollution, even at low concentrations in sediment, poses a serious threat to ocean environments [11]. Hence, heavy metals such as Cu, Cd, and Pb have been regarded as priority pollutants due to their highly toxic nature by various environmental protection agencies such as the US Environmental Protection Agency (USEPA) and World Health Organization (WHO). Research on the remediation of heavy-metal-contaminated dredged sediments has become more important. Among the multitude of disposal technologies of heavy metal remediation [12], chemical leaching has been applied in the field of actual contaminated soil and sediment remediation due to its advantages of high efficiency, thorough treatment, and strong adaptability. For example, data published by the USEPA showed that chemical leaching remediation has been used in more than 16 major contaminated site remediation projects in the past decade [13]. However, the leaching operation by using strong acids or chelating agents may destroy the physical and chemical properties of the dredged sediments, and there is also a risk of secondary pollution in using the leaching solutions [14].

The dredged sediments generated in most ports and waterways in China are contaminated by heavy metals to varying degrees [15], and the improper disposal of heavy-metal-contaminated dredged sediments directly or indirectly has a negative impact on marine ecosystems [16,17]. Natural organic acids are mildly acidic, biodegradable, and inexpensive compared to artificial chelating agents. The scientific hypothesis of this study is that natural organic acids have good application prospects in the leaching remediation of heavy-metal-contaminated dredged sediments [18,19]. The over-arching objective of this study was to use natural organic acids (oxalic acid, citric acid, tartaric acid, and malic acid) as leaching agents to remove heavy metals from contaminated dredged sediments. Batch experiments were conducted to investigate the factors governing the removal rate of heavy metals and leaching kinetics with different natural organic acids. The relevant findings provide technical support for the remediation and disposal of contaminated dredged sediments in ports and waterways.

2. Materials and Methods

2.1. Preparation of Contaminated Dredged Sediments

The surface sediment samples (~0–10 cm) were collected from the steel base operation area of the Yingkou Port (China, 40.29° N, 122.10° E). Triplicates (total 3.0 kg) were collected using a grab sampler with a capacity of 1.0 kg. These sediment samples remained moist, dark in color, dominated by fine-grained sediment, and belonged to clean dredged sediments. The scientific hypothesis is to remediate heavy-metal-contaminated dredged sediments. Therefore, to meet the experimental requirements, the typical heavy metals Cu, Cd, and Pb were used as target contaminants to prepare the heavy-metal-contaminated dredged sediments using the following method. The original dredged sediments (~1.0 kg) were sieved, placed in a plastic container, and added to a mixed solution (~1 L) of copper nitrate (400 mg/L), cadmium nitrate (10 mg/L), and lead nitrate (500 mg/L) to contaminate the dredged sediments. The contamination process lasted for 30 days of daily stirring, then the contaminated dredged sediments were dried in an oven (55 °C), homogenized, and ground.

2.2. Leaching Experiments

The leaching of Cu, Cd, and Pb in the contaminated dredged sediments by the natural organic acids was conducted in batch experiments (Table 1). All reagents used in the experiments were of guaranteed grade. Specifically, 0.5 g of the obtained contaminated dredged sediment was weighed in a 50 mL centrifuge tube, followed by shaking leaching using the organic acid solutions designed for the batch experiments. The reaction system was conducted in a constant temperature oscillator at a temperature of 25 °C. The mixtures were shaken for the pre-set time and then filtered immediately through 0.45 μm pore size membranes (Polyethersulfone, Jinteng, Tianjin, China). Those samples were stored at 4 °C in a dark location until they were tested. All experiments were carried out in triplicate to improve the statistic confidence of the experimental data.

Table 1. Experimental conditions.

	Experiment 1	**Experiment 2**	**Experiment 3**
Contaminated dredged sediments		0.5 g per experiment	
Organic acids	oxalic acid, citric acid, tartaric acid, and malic acid	oxalic acid, citric acid, tartaric acid, and malic acid	oxalic acid, citric acid, tartaric acid, and malic acid
Organic acid concentration (mmol/L)	0, 5, 10, 20, 30, 50	20	20
Solid-to-liquid ratio	1:20	1:10, 1:20, 1:30, 1:40	1:20
Leaching time (min)	20	20	0, 5, 10, 20, 30, 60

2.3. Analytical Methods and Data Processing

The dredged sediment mineral characterization was performed using XRD (D8Advance, Bruker), and then the d value and the corresponding relative intensity I value of each diffraction line were calculated. Finally, JCPDS cards were compared to determine the mineral classification of each peak. The particle size distribution of the dredged sediment was determined with a laser particle sizer (Bettersize3000Plus, Dandong, China). The pH and conductivity of the dredged sediment were determined in a mixture of sediment:Milli-Q water at a ratio of 1:2.5 suspensions with a METTLER TOLEDO pH meter and an MC226 Basic Conductivity Meter. The cation exchange capacity of the dredged sediment was analyzed according to the determination of the cation exchange capacity and the exchangeable base of neutral soil (Y/T 295-1995, China). The total heavy metal content of the dredged sediment was analyzed at the Shiyanjia Lab (www.shiyanjia.com). The samples were digested and processed according to Pretreatment Guideline of Heavy Metals Analysis in the Marine Sediments and Organisms-Microwave Assisted Acid Digestion (HY/T 132-2010, China), and the Cu, Cd, and Pb were measured with inductively coupled plasma optical emission spectrometry (ICP-OES).

The heavy metals in the leaching solution were determined using ICP-OES (ICP-5000, Focused Photonics inc., Hangzhou, China) with a detection limit of 1 μg/L for Cu, Cd, and Pb, stability $\leq$1.0% @ 2 h, and precision $\leq$1.0% [20]. To improve the quality of the experimental data, we conducted three replicate experiments with two times the standard deviation as the error. In this study, the removal rate (%) of heavy metal by organic acid leaching and the amount of heavy metal leached per unit mass of sediments (q, mg/Kg) were calculated using Equations (1) and (2), respectively:

$$\text{removal rate } (\%) = \frac{C_l V_l}{C_s m_s} \times 100\% \quad (1)$$

$$q = \frac{C_l V_l}{m_s} \quad (2)$$

where C_l (mg/L) and C_S (mg/kg) are the concentrations of heavy metal in leaching solution and dredged sediment, respectively; V_l is the volume of leaching solution (L); and m_S is the mass of the sediment (Kg).

3. Results and Discussion

3.1. Characterization of the Initial Dredged Sediments

Defining the composition as well as characterizing the physicochemical properties of the dredged sediments is therefore regarded as an essential step towards identifying suitable remediation methods and future beneficial uses [1]. The XRD results (Figure 1) showed that the initial dredged sediment was composed of Quartz [SiO_2], Dolomite [$CaMg(CO_3)_2$], Mica [$KAl_2(Si_3Al)O_{10}(OH,F)_2$], Kaolinite [$Al_2Si_2O_5(OH)_4$], Tremolite [$Ca_2Mg_5(Si_4O_{11})_2F_2$], Calcite [$CaCO_3$], and Albite [$(Na,Ca)Al(Si,Al)_3O_8$]. The basic characteristics of the dredged sediment are listed in Table 2. The predominant fraction of the dredged sediment was fine particles $\leq$63 μm, and the silt and clay proportions were 52% and 31%, respectively. The pH, electrical conductivity, and cation exchange capacity of the dredged sediment were 7.47, 1.21 mS/cm, and 9.68 cmol/kg, respectively. The total contents of Cu, Cd, and Pb in the initial collected dredged sediments were 42.1, 0.3, and 54.7 mg/Kg, respectively, and their concentrations increased to 378.0, 6.9, and 489.0 mg/kg for the test dredged sediments after the contamination operation, which were significantly above the tolerable limits recommended by marine sediment quality (GB 18668-2002, China). The fine-grained dredged sediments in this study are categorized as silt and clay which are more associated with contamination compared with sandy sediments in other ports since clay and silt easily bind with pollutants [21]. In addition, the electrical conductivity and cation exchange capacity are relatively high for the dredged sediments indicating a high ionic charge on their surfaces, and it has been shown that the extraction of heavy metals with organic solutions (e.g., natural organic acids) requires an environment with a complex ionic charge when submitted to the leaching process [22]. The literature studies also show that most of the heavy metals in sediments exist as cationic species, and the cations of these metals can compound with inorganic components in sediments to form precipitation or positively charged complexes [23]. Accordingly, leaching remediation of the heavy-metal-contaminated dredged sediment using natural organic acids is a suitable and promising choice.

Table 2. Basic features of the initial collected dredged sediment.

	Particle Composition	pH	Conductivity	Cation Exchange Capacity	Total Heavy Metals (mg/kg)		
			(mS/cm)	(cmol/kg)	Cu	Cd	Pb
Dredged sediment	Silt (<63 μm): 52% Clay (< 2 μm): 31%	7.47	1.21	9.68	42.1	0.3	54.7

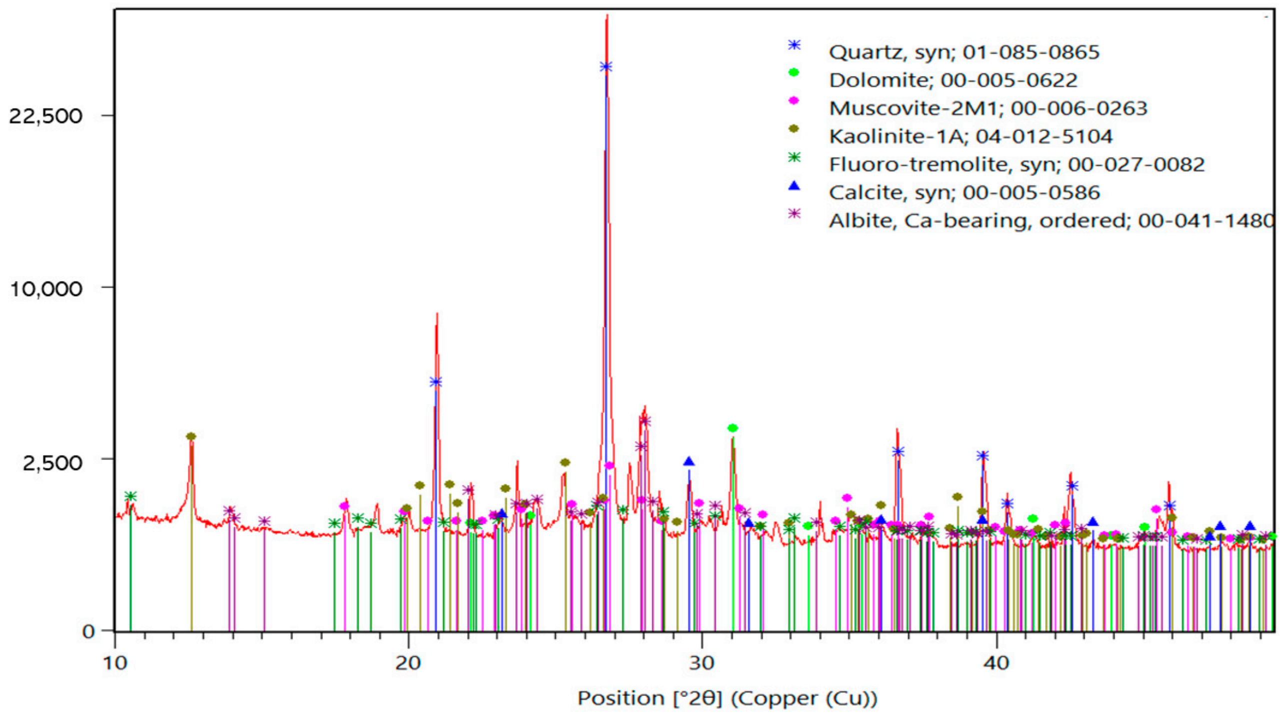

Figure 1. XRD patterns of marine dredged sediments.

3.2. Factors Governing the Leaching Efficiency of Heavy Metals by Natural Organic Acids

Influence of the organic acid and its concentration: Acid leaching is a common method for removing heavy metals from contaminated sediments, and among which, natural organic acid is considered as a promising leaching agent because it is biodegradable, yielding little damage to the sediments with a low risk of secondary pollution [24,25]. The contaminated dredged sediments were leached with various concentrations of natural organic acids to investigate their effects on heavy metal removal. The results of the leaching experiments (Figure 2) showed that with an increase in the organic acid concentration, the removal of Cd, Cu, and Pb gradually increased and then tended to stabilize. The concentration of the organic acids was the key factor in the reaction. An important contribution of the organic acid leaching mechanism is the formation of highly soluble complexes between organic acid functional groups and heavy metals [14]. As the concentration of the leaching agent increases, the number of functional groups participating in the reaction is saturated for heavy metals, and therefore the leaching efficiency gradually plateaus [26]. Other explanations might be attributed to the Fe^{3+} in the sediments hindering the ability of the leaching agents to bind to the target heavy metals [27]. The results indicate that the concentration needs to be appropriate for the actual project due to the fact that high concentrations of leaching agent could lead to an increase in costs.

Oxalic acid showed a good leaching effect on Cd, with a maximum removal of 62%, while only approximately 20% of Cu and Pb were removed. Citric acid had the best leaching effect on all three heavy metals, removing greater than 80% at a concentration of 20 mmol/L. Tartaric acid had a good leaching effect on Cd and Cu, removing close to 80% at a concentration of 20 mmol/L, while its effect on Pb was not significant. Malic acid also performed well in the leaching of Cd, Cu, and Pb, and its removal rate of heavy metals reached the highest level at a concentration of 20 mmol/L, but the overall performance was lower than that of citric acid. The leaching effects of the different organic acids on the removal of Cu, Cd, and Pb from the test dredged sediments were ranked in descending order as citric acid > malic acid > tartaric acid > oxalic acid. Therefore, citric acid had the best leaching effect on Cd, Cu, and Pb, with an optimal leaching concentration of 20 mmol/L. The difference in removal efficiencies might be ascribed to different functional groups in the organic acids that could bind heavy metal ions to different degrees [28]. Citric acid has an increased number of carboxyl compared to other acids, which could bind heavy metal ions more easily. The findings are in accordance with the results of a study conducted by Geng et al. [29] which indicate that citric acid preferably leached metal ions.

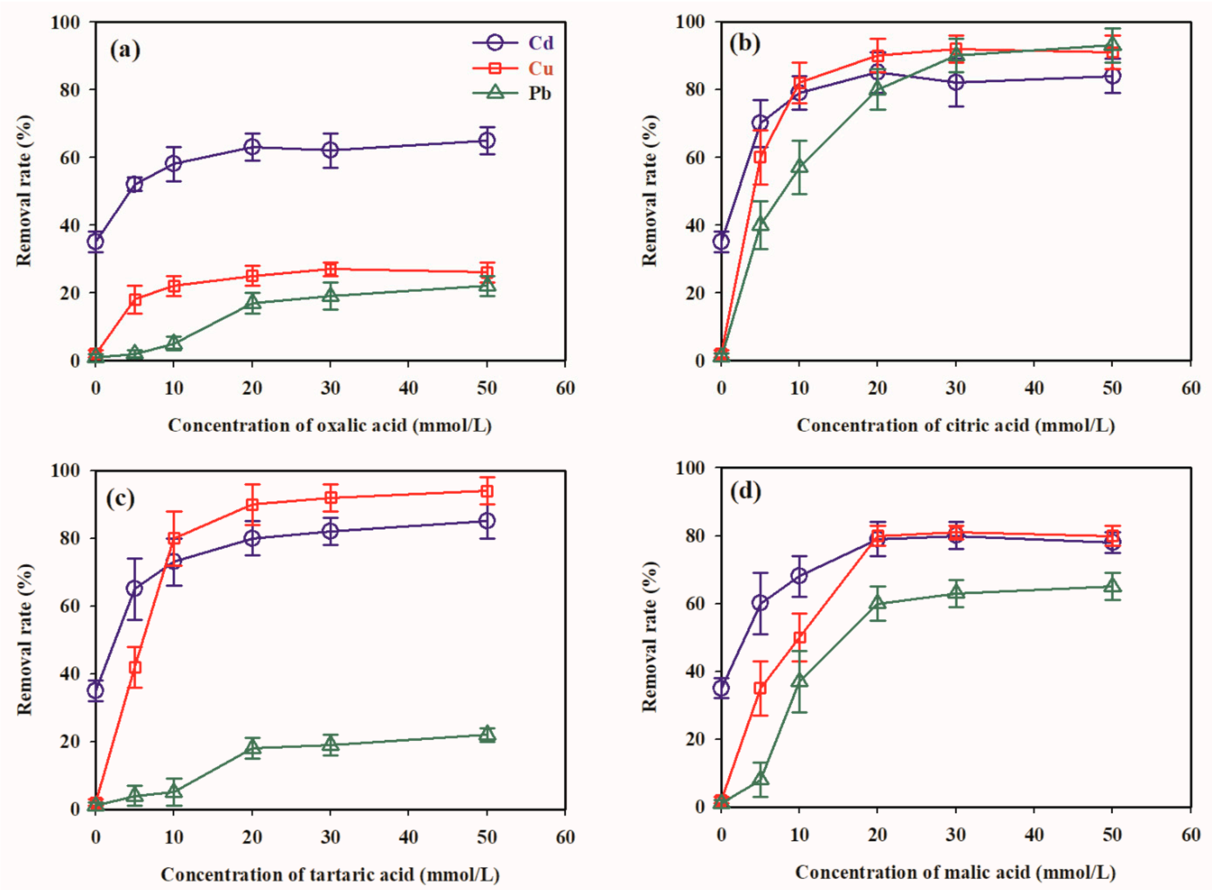

Figure 2. Effect of organic acid concentration on the removal of heavy metals from contaminated dredged sediments ((**a**), oxalic acid; (**b**), citric acid; (**c**), tartaric acid; (**d**), malic acid).

Influence of the solid-to-liquid ratio (SLR): The SLR is the ratio of the mass of the contaminated dredged sediment remediated by leaching to the volume of the leaching agent and can directly influence the effect and cost of leaching [30]. The results of this study showed that as the SLR gradually decreased from 1:10 to 1:40, the leaching effect of organic acids on the heavy metals Cu, Cd, and Pb first showed an increasing and then stabilizing trend (Figure 3). As the SLR decreased, the content of organic acids that can participate in the desorption and chelation of Cu, Cd, and Pb increased [31], and therefore, their relative leaching efficiency increased. The decreasing SLR led to a decrease in the diffusion resistance, which in turn led to an increase in the interaction of sediments and acid during leaching [32]. A lower SLR increases the amount of organic acids available, but may also decrease the heavy metal concentration in the leaching solutions. A concentration of heavy metals in the leaching solutions that is too low is detrimental to the leaching process [33]; hence, an SLR of 1:20 is appropriate.

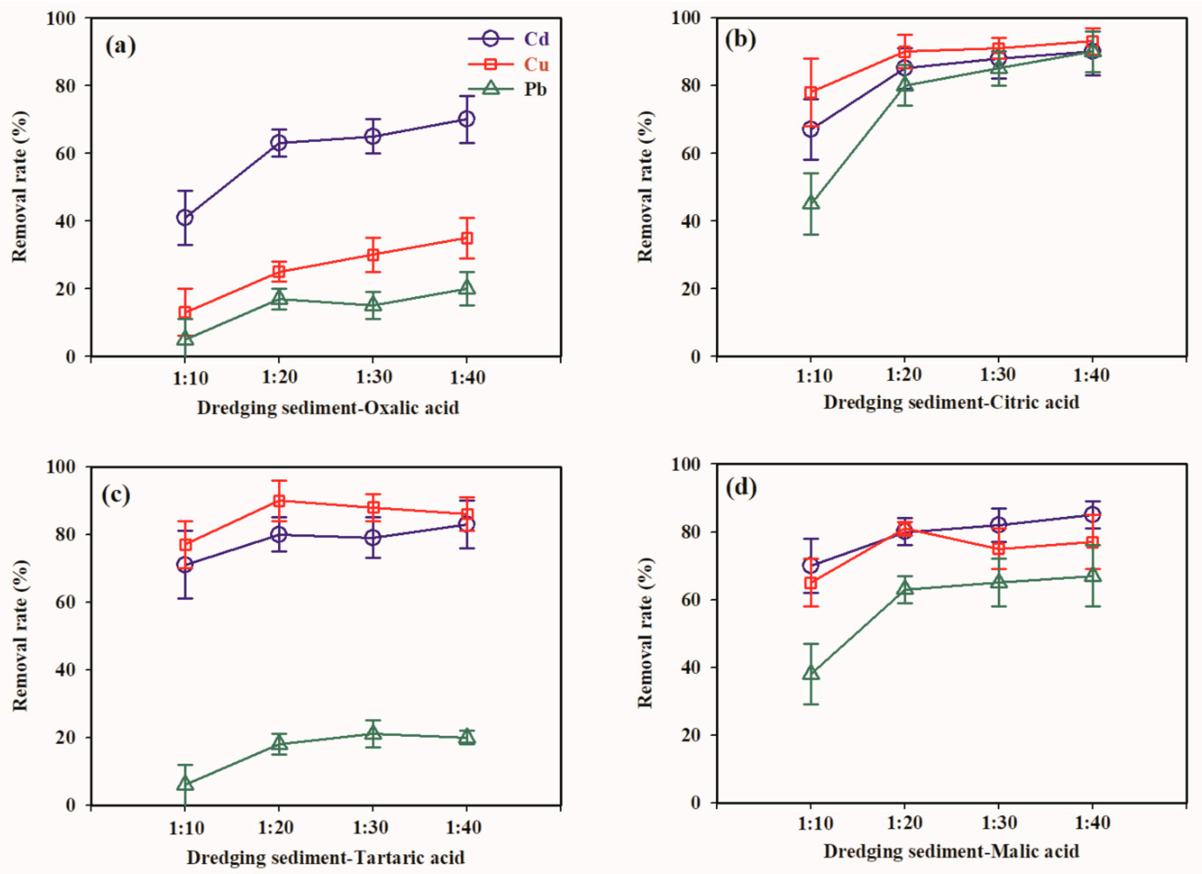

Figure 3. Effect of the solid-to-liquid ratio on the removal of heavy metals from polluted dredged sediments ((**a**), oxalic acid; (**b**), citric acid; (**c**), tartaric acid; (**d**), malic acid).

3.3. Kinetics of Leaching by Natural Organic Acid

The evaluation of the leaching kinetics can provide important information about the interactions between heavy metals, organic acid, and dredged sediment [23]. The kinetics of the leaching of Cu, Cd, and Pb from the contaminated dredged sediment by citric acid was studied, and the results can be seen in Figure 4a. It is clear that the removal of heavy metals increased rapidly with the leaching time initially and then gradually reached the maximum value. The leaching of Cu from the contaminated dredged sediment increased with the leaching time and began to balance after 20 min with the removal rate reaching 90% (37.9 mg Cu/kg dredged sediment). A similar trend was observed for Cd and Pb, while the equilibrate removal rate reached 85% (0.9 mg Cd/kg dredged sediment) and 80% (43.8 mg Pb/kg dredged sediment) after 20 min of leaching, respectively. The leaching process makes the heavy metals continuously adsorb and desorb between the dredged sediments and the leaching agent, eventually reaching a dynamic balance [34].

There are many theoretical models that could serve to understand the dynamics of leaching processes. In this study, the experimental results were analyzed using the Elovich equation [35] and the two-constant rate equation [36]. The Elovich equation describes a series of reaction mechanisms and is commonly used to describe the chemical kinetics of heavy metal adsorption and desorption at the interface of sediments. The Elovich equation is expressed as follows:

$$q = (1/\beta_s)ln(\alpha_s\beta_s) + (1/\beta_s) \times ln(t) \tag{3}$$

where q is the amount of heavy metals removed from the test dredged sediments; t is the time; α_s is the initial adsorption rate; and β_s is the desorption coefficient, respectively.

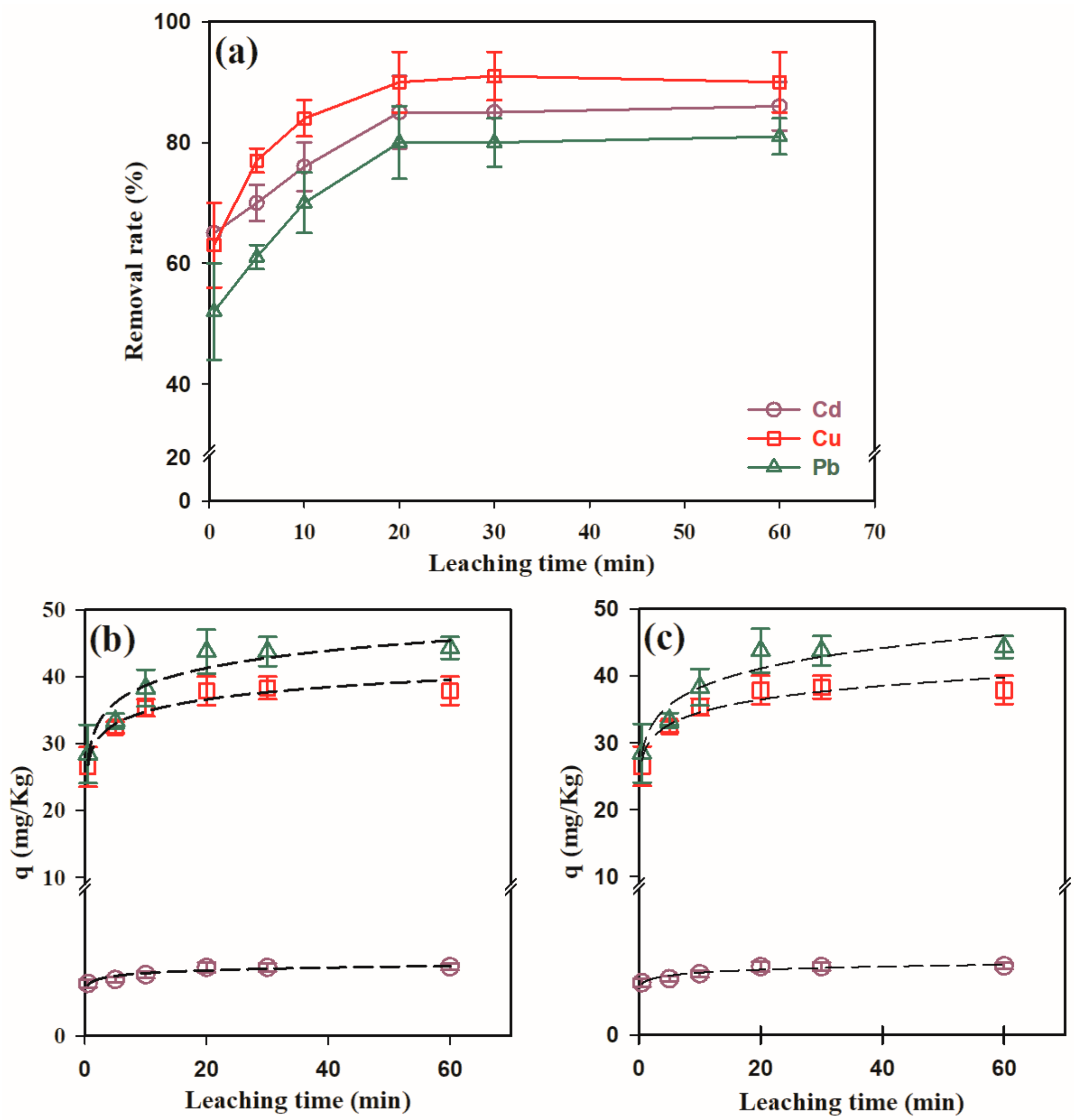

Figure 4. (**a**) Effect of the leaching time on the removal rate of heavy metals from contaminated dredged sediments by citric acid; (**b**,**c**) are Elovich equation and two-constant equation kinetics models plots for the leaching of Cu, Cd, and Pb by citric acid, respectively.

The two-constant rate equation is the modified Freundlich equation and is suitable for describing the kinetics of heavy metal adsorption and desorption with a complex reaction process. The double constant equation is expressed as follows:

$$q = a \cdot t^b \quad (4)$$

where q is the amount of heavy metals removed from the test dredged sediments; t is the time; a is the initial metal desorption rate constant; and b is the desorption rate coefficient.

Kinetic models are shown in Figure 4b,c. The Elovich and two-constant rate models converge well with acceptable regression coefficients which indicates the suitability of the two kinetic models for the description of the leaching of heavy metals from contaminated dredged sediment by citric acid. The model parameters were calculated and are summarized in Table 3. It was found that the Elovich equation results had a much better fit with correlations at significant levels indicating that the kinetics of remediating heavy-metal-contaminated dredged sediments by natural organic acid leaching is a heterogeneous diffusion process. The same observations were found compared with other literature reports, e.g., Cu desorption of calcareous soils [37] and Cd desorption from sub-tropical soils [38]. These results indicated that the leaching remediation process by organic acid depended on the sediment properties as well as the heavy metal species in sediments.

Table 3. Kinetic model parameters for the leaching of Cu, Cd, and Pb from dredged sediments.

Heavy Metal	Elovich Equation			Two-Constant Rate Equation		
	α_s	β_s	R^2	a	b	R^2
Cu	359	0.329	0.9931	27.9	0.09	0.9805
Cd	5.3	1.667	0.9911	0.7	0.07	0.9843
Pb	152	0.229	0.9943	28.9	0.12	0.9891

4. Conclusions

The contaminated dredged sediments in ports and waterways should be decontaminated and disposed of according to their contamination level prior to dumping or resource utilization. This study investigated the pattern of removing heavy metals from dredged sediments by natural organic acid leaching. The leaching effects of organic acids were ranked in descending order as citric acid > malic acid > tartaric acid > oxalic acid. Citric acid had the best leaching effect on heavy metals Cu, Cd, and Pb from contaminated dredged sediments, with an optimum organic acid concentration of 20 mmol/L and an optimum SLR of 1:20. The leaching kinetics was best fitted using the Elovich equation, indicating that the remediation of heavy-metal-contaminated dredged sediments by natural organic acid leaching is a heterogeneous diffusion process. The contaminated dredged sediments could be remediated by using the citric acid leaching process effectively and successfully within a short period of time which can provide technical support for the remediation and disposal of contaminated dredged sediments in ports and waterways.

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