Long-term effects of low-molecular-weight organic acids on remobilization of Cd, Cr, Pb, and As in alkaline coastal wetland soil

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ABSTRACT

Low-molecular-weight organic acids (LMWOAs) that have a wide variety of biological activities can affect the mobilization of heavy metals in soil. In this study, the effects of three kinds of LMWOAs on the remobilization of Cd, Cr, Pb, and As were investigated. The results showed that the concentrations of Cr, Pb, and As reached the maximum values at 7 d, and then decreased at 58 d, indicating that the LMWOAs induced the release of these elements from soil. The ability of LMWOAs to affect the mobility of Cr and As followed the order citric acid (1.32 and 2.51mg/L) > malic acid > oxalic acid, while the order for Pb was malic acid (4.49mg/L) > citric acid > oxalic acid. In addition, the sequential extraction indicated that the LMWOAs advanced the reactivation of Pb, Cd, and As, but led to the immobilization of Cr after a long-term reaction.

1. Introduction

In recent years, natural and human activities have a great impact on the environment, resulting in increasingly greater heavy metal pollution in soil, sediment, and surface water [1,2]. Coastal wetland soil suffers from heavy metal pollution globally, which has become an important environmental health problem in the world [3,4]. Heavy metals, such as Cr, Pb, and Cd, do great harm to ecosystems due to their potential reactivity, mobility, and toxicity, which can be continuously accumulated in wetland soil [5,6]. They can transfer from soils to plants, accumulate in plant tissues, and further affect the ecological environment and human health [7].

Low-molecular-weight organic acids (LMWOAs) are ubiquitous water-soluble compounds that participate in many biological processes and are important intermediates in the cycle of tricarboxylic and glyoxylic acid [8–10]. LMWOAs are excreted by plant roots, and the activity of LMWOAs affects the transport and toxicity of heavy metals in soils [11–15]. LMWOAs can react with heavy metals in complexation, precipitation, and redox, and therefore induce to the adsorption and release of heavy metals from the solid, which can affect the solubility and bioavailability of heavy metals. Therefore, many studies have focused on the impact of LMWOAs on the mobilization of As with the participation of microorganisms in mangrove wetlands [24,25]. The microbe facilitated As extraction of citric and malic acid in mangrove sediments [25]. At present, the previous researches primarily focus on the impact of LMWOAs on the mobilization of heavy metals in acid soils, and the reaction time being only several days [23,26–29], which is not sufficient for evaluating LMWOAs-driven mobilization of heavy metals in various soil types. Few studies, to the best of our knowledge, have concentrated on the influence of LMWOAs on heavy metals mobility in alkaline wetland soils with a long time [30]. Therefore, to further understand the long-term influence of LMWOAs on the mobility of heavy metals in alkaline wetland soils, the objectives of the present study are the following: (1) compare the influence of three kinds of LMWOAs (citric, oxalic, and malic acid) and the mixed acids of LMWOAs on the mobility of Pb, Cd, Cr, and As in alkaline wetland soils over a period of approximately 2 months, and (2) study how the LMWOAs affect the fractionation of Pb, Cd, Cr, and As in alkaline wetland soils using the sequential extraction method.

2. Materials and methods

2.1 Soil samples

Two wetland alkaline soil samples (soil SDZZ-2 and soil P6-2) were taken from the Yancheng tidal flat wetland in Jiangsu Province along the coastline of the Yellow Sea in eastern China. The Yancheng tidal flat wetland is
the largest typical muddy tidal flat wetland in China with the most complete ecological types. In recent years, the tidal flat wetland has undergone contamination with heavy metals, such as Pb, Cd, Cr, and Zn, due to great economic development including reclamation, fish and shrimp farming, and industrial construction [31]. Soil samples were collected from the upper layer (0–20 cm). The samples were placed in polypropylene bags, stored in coolers, and transferred to the laboratory. The soil samples were air-dried, then sieved through 2 mm nylon sieves. The chemical characteristics of the soil samples are shown in Table S1.

### 2.2 Batch experiments

Three representative LMWOAs, including malic, citric, and oxalic acid, were selected and seven treatments were designed. The different treatments designated A1, A2, A3, A4, A5, A6, and A7 were performed separately, as shown in Table 1.

After 2 g of the soil sample was mixed with 25 ml of LMWOA solution in a 50 ml plastic centrifuge tube according to the designed treatment method (Table 1), each centrifuge tube was shaken for approximately 1 min, and then the centrifuge tube was placed into the tube rack at room temperature. The batch experiments were conducted for 58 d, and the centrifuge tubes were taken at 7 d, 28 d, and 58 d, respectively. The pH values and the electrical conductivity (EC) of the samples were monitored over the experimental period. The suspension samples in centrifuge tubes were separately collected, and then centrifuged (4000 g, 8 min, 25°C). The supernatants were passed through a 0.22 μm syringe filter for analysis of dissolved Pb, Cd, Cr, As, and Fe. The centrifuged solid samples at 28 and 58 d were collected and used for the sequential extraction. In this study, Fe was also analyzed because that Fe oxides are the predominant binders for heavy metals in soils [23,32].

### 2.3 Sequential extraction procedure

Sequential extraction procedures are commonly applied to estimate the bioavailability of heavy metals in soils and sediments [33,34]. The sequential extraction procedures were applied to 1.000 g of centrifuged solid sample at 28 d and 58 d using different reagents, and experimental conditions were summarized in Table S2. Details of the sequential extraction procedures can be found elsewhere [35,36]. The extraction solution obtained from each step of the sequential extraction was filtered by a 0.22 μm syringe filter for analysis of Pb, Cd, Cr, As, and Fe concentrations.

#### 2.4 Aqueous analysis

The concentrations of Pb, Cd, Cr, As, and Fe in the solution were measured by an atomic absorption spectrometer (AAS-TAS990, Beijing Pu-Xi, China). For quality control, external standards and spiked samples were tested to ensure the accuracy of the chemical analyses. The recovery range was 88–110%. Soil pH was measured using a pH meter (pHS-3 C, China), and EC was measured using a conductivity meter (DDSJ-318, China).

## 3. Results and discussion

### 3.1 Solution pH in various treatments

The pH values of solutions with the seven treatments (Table 1) for the two soil samples (soil SDZZ-2 and soil P6-2) are presented in Figure 1. The range of pH values was between 2.21 and 8.14 for the seven treatments. In general, the pH value was low initially, then increased continuously with reaction time, and finally reached stability (58 d). For the soil SDZZ-2 and soil P6-2, the tendency of the pH value was similar with seven treatments. The pH values of soil P6-2 were higher than that of soil SDZZ-2 before 28 d with the seven treatments, and these values were basically the same at 58 d in two soils. In addition, for the seven treatments, the EC values were high initially (mean 2.40 ± 0.70 ms/cm), then decreased continuously with reaction time, and reached 1.31 ± 0.31 ms/cm in average at 58 d with soil SDZZ-2 and soil P6-2 (Fig. S1).

### 3.2 Pb, Cd, Cr, and As in solutions on day 7 in various treatments

Figure 2 shows the concentrations of Pb, Cd, Cr, As, and Fe in solutions for soil SDZZ-2 and soil P6-2 with seven treatments after 7 d of reaction. For the single-acid treatments (A1, A2, and A3), the tendencies of Cr, Cd, As, and Fe concentrations in the two soil samples were similar. The ability of single LMWOAs to affect the mobility of Cr, Cd, and As followed the order citric acid > malic acid > oxalic acid. The concentrations of Cr, As, and Fe were the highest (1.32, 2.42, and 1193.18 mg/kg, respectively) with citric acid (A1), followed by malic acid (A3), and the lowest with oxalic acid (A2) in soil SDZZ-2. The concentrations of Cr, As, and Fe also reached the highest values (1.31, 2.51, and 1632.84 mg/kg) with citric acid (A1), followed by

### Table 1. Seven different organic acids treatments.

| Treatment | Solutions (0.01 M) |
|-----------|--------------------|
| A1        | Citric acid        |
| A2        | Oxalic acid        |
| A3        | Malic acid         |
| A4        | Oxalic acid        |
| A5        | Malic acid         |
| A6        | Malic acid         |
| A7        | Citric acid        |
|           | Malic acid         |
malic acid (A3), and the lowest values with oxalic acid (A2) in soil P6-2. Our results were consistent with the previous findings that the concentration of extracted Cr in contaminated soil increased when the LMWOAs were substituted in this order: citric acid > malic acid > oxalic acid [32]. The Cd concentrations were not detected in the most of seven treatments, however, reached the highest value (0.004 mg/kg) with citric acid (A1) in soil SDZZ-2, and the highest value (0.013 mg/kg) with mixed acid (citric, malic, and oxalic acid, A7) in soil P6-2. However, the degree of influence by LMWOAs for Pb is different from the others, and followed the order malic acid > citric acid > oxalic acid. The concentrations of Pb reached the maximum values of 3.79 and 4.49 mg/kg in soil SDZZ-2 and soil P6-2, respectively, with malic acid (A3), and were lower with citric acid (A1) and oxalic acid (A2).

For the mixed-acid treatments (A4, A5, A6, and A7), in most cases, the concentrations of Cr, Cd, As, and Fe in solutions were less than those treated with A1 (citric acid) in the two soil samples, except for Cd and Cr in soil P6-2. The most interesting observation was that the concentrations of Cr, Cd, As, and Fe in this study with the mixed-acid treatments were lower than the sum of heavy metal concentrations in the single-acid treatments (Figure 2). Therefore, the effect of mixed-acid treatments on the mobility of Cr, Cd, and As was not obvious compared with single-acid treatments, which indicates that there

Figure 1. Solution pH values in soil SDZZ-2 and soil P6-2 with seven treatments (A1, A2, A3, A4, A5, A6, and A7).
was no significant ‘superposition effect’. However, the concentrations of Pb reached the maximum values of 5.20 and 5.54 mg/kg with A5 (citric and malic acid) treatment in soil SDZZ-2 and soil P6-2, respectively.

The chemical reactions of organic acids dissolving heavy metals in soils mainly include acidification, reduction, and complexation [15,23,37]. In this study, the effect of citric, oxalic, and malic acid on Pb, Cd, Cr, and As is different, and, in most cases, the degree of influence is citric acid > malic acid > oxalic acid for Cd, Cr, and As. Previous studies also reported that organic acids with different carboxylic groups have different abilities for releasing heavy metals [32,38]. The activation of tricarboxylic (citric) acid is stronger than that of dicarboxylic (malic and oxalic) acid, because of it has a larger molecular weight and surface area, carries more negative charges, and possesses a stronger ability to adsorb or chelate heavy metals [30,39]. Moreover, the concentrations of Pb were different from that of other heavy metals. The higher concentrations of Pb were in treatments of A3 (malic acid), A5 (citric and malic acid), and A7 (oxalic, citric, and malic acid) at 7 d, and the three treatments all included malic acid, which indicates that the effect of malic acid on Pb is more greater than other LMWOAs [37,40] in the initial stage of reaction. In accordance with our results, reported that citric acid promoted Cd and Zn mobility in soils but only little influence on Pb, which might be due to the low affinity of citric acid to Pb.

### 3.3 Pb, Cd, Cr, and As in solutions on day 28 and 58 in various treatments

The concentrations of Pb, Cd, Cr, As, and Fe in solutions after 28 and 58 days of reaction are shown in Figures 2 and 3. In general, for single-acid treatments (A1, A2, and A3), the concentrations of Pb, Cr, and Cd in the solution at 28 d were the highest with citric acid (A1) and the
lowest with oxalic acid (A2) in the two soil samples. However, the concentrations of As and Fe reached the highest values of 0.58 and 0.07 mg/kg with malic acid (A3) in soil P6-2. Moreover, the effects of mixed-acid treatments (A4, A5, A6, and A7) on the mobility of Pb, Cd, Cr, and As were also no significant ‘superposition effect’, which were similar with the 7 d reactions.

On day 58, the range of Pb, Cr, and As concentrations was 0.004–0.2, 0.01–0.05, and 0.1–0.4 mg/kg in soil SDZZ-2, while that was 0.004–0.1, 0.006–0.007, and 0.2–1.0 mg/kg in soil P6-2 with seven treatments, respectively (Figure 3). There was no significant tendency of heavy metals among the seven treatments. However, it is worth noting that the concentrations of

Figure 3. Concentrations of Cd, Cr, Pb, As, and Fe in solution at 7, 28, and 58 d in soil SDZZ-2 and soil P6-2 with seven treatments (unshown denotes undetected).
Pb, Cr, As, and Fe generally decreased at 58 d compared with that at 7 d, except for Cd, which was different from previous studies [23].

The pH values can affect the mobility of heavy metals and existence form of LMWOAs in solutions [38,41]. The low pH is conducive to the dissociation of heavy metals from the soil, and LMWOAs ligands can react with heavy metals to form soluble binary chelates through functional groups such as carboxyl and hydroxyl [38,42], which led to the release of heavy metals with high concentrations at 7 d (Figures 1 and 2). However, as time went on, LMWOAs reacted with various soil components at different rates, and the proton in the solution was increasingly consumed until all possible reactions reached equilibrium [5,23], which resulted in an increase in solution pH to about 8 at 58 d (Figure 1). The availability and complexation effect of LMWOAs ligands decreased, causing the decrease of Pb, Cr, and As concentration in solution at 58 d (Figure 3). In addition, under alkaline conditions (high pH), organic acids are dissociated and form more anions, which can form ternary surface complexes with heavy metals on soil surface. Meanwhile, the increased pH can promote the precipitation of partially dissolved heavy metals. These were another possible reasons for the decrease of Pb, Cr, and As concentrations at 58 d.

However, the effect of pH cannot entirely explain the phenomenon observed in this study, such as Cd. The concentration of Cd was little at 7 d (low pH) (Figures 1 and 2), and increased significantly at 28 d and 58 d (high pH) (Figures 1 and 3). The high concentrations of Cd after 28 d and 58 d may be due to the following reasons: (1) the initial content of heavy metal in soils is one of the factors affecting mobility of heavy metals [43,44], and the content of Cd in the original soil is low in this study (Table S1); (2) the properties of soil are also one of the important factors affecting the mobility of heavy metals [32]; and (3) it possibly requires more time to allow reaction between the LMWOAs and Cd for the release of Cd from the soils.

### 3.4 Correlation analysis

According to the previous studies, Fe oxides are the predominant binders for heavy metals in soils [23,32], therefore, the correlation analyses between Fe and Cr, Pb, As, and Cd concentrations after 58 d are shown in Figure 4. The results demonstrate that the correlations between Fe and Cr, Pb, and As were all significant. The correlation coefficient ($R^2$) of Cr was 0.72 ($p < 0.05$) and 0.67 ($p < 0.05$) in soil SDZZ-2 and soil P6-2, respectively.

![Figure 4](image-url). Linear relationship between concentrations of Fe and Cd, Cr, Pb, and As after 58 d in soil SDZZ-2 and soil P6-2 with seven treatments.
The $R^2$ of Pb was 0.70 ($p < 0.05$) and 0.79 ($p < 0.05$) in soil SDZZ-2 and soil P6-2, respectively. Meanwhile, there was a significant correlation between As and Fe, and the $R^2$ was 0.55 ($p < 0.05$) and 0.48 ($p < 0.05$) in soil SDZZ-2 and soil P6-2, respectively. The results indicate that the LMWOA-driven release of Cr, Pb, and As in the two soil samples is significantly affected by mobilization of Fe oxides.

In addition, to verify the effect of pH in the LMWOA-driven mobility of heavy metals, the correlation analyses between pH and Cr, Pb, As, Fe, and Cd after 58 d are presented in Figure 5. There was a good relationship between the pH and Fe in soil SDZZ-2 ($R^2 = 0.84$, $p < 0.05$) and soil P6-2 ($R^2 = 0.82$, $p < 0.05$). The pH values also had significant relationships with Pb and As in soil SDZZ-2 ($R^2 = 0.61$, $p < 0.05$; $R^2 = 0.53$, $p < 0.05$) and P6-2 ($R^2 = 0.70$, $p < 0.05$; $R^2 = 0.51$, $p < 0.05$). In agreement with our results, 23 also reported that As and Pb had significant relationships with pH in soil solutions with the addition of LMWOAs. Meanwhile, the pH value was correlated with Cr in soil SDZZ-2 ($R^2 = 0.63$, $p < 0.05$) and soil P6-2 ($R^2 = 0.37$, $p < 0.05$). The pH was significantly correlated with Fe, Pb, As, and Cr, which further proves that pH had a strong influence on the release of these heavy metals from soils in the coastal alkaline wetland environment. However, Cd had no significant relationship with pH (Figure 5) and Fe (Figure 4) in this study, which suggest that the release of Cd in the system was not only controlled by pH and Fe oxides.

### 3.5 Fractionation of Pb, Cd, Cr, and As in soils

A sequential extraction method was applied to analyze the chemical forms of Pb, Cd, Cr, and As in the two soils. The results of the fractionation analyses with seven treatments are shown in Figures 6 and 7. The LMWOAs in seven treatments all resulted in the remobilization of Pb, Cd, Cr, and As in soil SDZZ-2 and soil P6-2. Compared with the raw soil, the exchangeable and carbonate-bound Pb, and Fe-Mn oxide bound Pb were all increased at 28 d and 58 d (Table 2, Fig. S2). At 28 d, the percentages of the exchangeable and carbonate-bound Pb, Fe-Mn oxide bound Pb, sulphides and...
Figure 6. Sequential extraction of Cd, Cr, Pb, As, and Fe in soil SDZZ-2 at 28 d and 58 d with seven treatments. (F1: exchangeable and carbonate-bound, F2: Fe-Mn oxide bound, F3: sulphides and organics bound, F4: residual).

organics-bound Pb, and residual Pb were 1.50%, 26.00%, 14.89%, and 57.62%, and 1.49%, 46.21%, 18.76%, and 33.54% in soil SDZZ-2 and soil P6-2 with seven treatments, respectively (Table 2, Figure 6). At 58 d, the exchangeable and carbonate-bound Pb, Fe-Mn oxide bound Pb increased to 4.57% and 30.14% in soil SDZZ-2 and 5.30% and 40.91% in soil P6-2, while the residual Pb decreased to 57.47% and 41.19% in soil SDZZ-2 and soil P6-2 with seven treatments, respectively. The tendencies of Cd and As fractionation were similar with Pb. The exchangeable and carbonate-bound Cd, Fe-Mn oxide bound Cd increased to 28.63% and 51.11% in soil SDZZ-2, and 25.41% and 66.69% in soil P6-2 at 28 d. At 58 d, the two values were 46.95% and 37.73% in soil SDZZ-2, and 50.96% and 40.33% in soil P6-2 with seven treatments. The
exchangeable and carbonate-bound As, Fe-Mn oxide bound As also increased to 0.93% and 3.96% at 28 d, and 1.26% and 5.04% at 58 d in soil SDZZ-2 with seven treatments.

However, the tendency of Cr fractionation was different from Cd, As, and Pb. Compared with the raw soil, the exchangeable and carbonate-bound Cr, Fe-Mn oxide bound Cr increased at 28 d, but decreased at 58 d in the two soils (Table 2, Figures 6, and 7). At 28 d, the percentages of the exchangeable and carbonate-bound Cr, Fe-Mn oxide bound Cr, and residual Cr were 0.60%, 1.58%, and 94.91% in soil SDZZ-2, and 0.52%, 1.59%, and 94.94% in soil P6-2 with seven treatments, respectively. At 58 d, the exchangeable and carbonate-bound Cr, Fe-Mn oxide bound Cr decreased to 0.22% and 0.08% in soil SDZZ-2, and 0.17% and 0.08% in soil P6-2, while the residual Cr increased to 97.43% and 97.28% in the two soils. For Fe, compared with the raw
Heavy metals in exchangeable and carbonate-bound, and Fe-Mn oxide bound have relatively high mobility and bioavailability. When the LMWOAs solution was added to the soil, the heavy metals in the acid extractable state were largely released due to the acid solubility of LMWOAs. The increase of Fe-Mn oxide bound fraction may be due to the chelation of LMWOAs [38]. The pH of the solution is an important factor affecting the fractionation of heavy metals. Forty-four, reported that the exchangeable and carbonate-bound, Fe-Mn oxide bound fractions were positively correlated with pH values. In this study, the pH value of soil solution increased to approximately 8 after Sd d of reaction (Figure 1), and the increased exchangeable and carbonate-bound, and Fe-Mn oxide bound fractions of Cd, As, Pb, and Fe were also observed in the two soil samples. However, the Fe-Mn oxide bound fraction of Cr and Fe all decreased at Sd d compared with the raw soil after LMWOAs addition, and the residual fractions of these two elements increased. The reason may be that there was formation of precipitate which resulted in the immobilization of Cr with Fe oxides in soils. In addition, the effect of different LMWOAs treatments (A1, A2, A3, A4, A5, A6, and A7) on the fractionation of Cd, Cr, Pb, and As was no obviously different.

4. Conclusions

The present study reveals the specific effects of low-molecular-weight organic acids (LMWOAs) on the mobility of Cd, Cr, Pb, and As in alkaline wetland soils. In the natural environment, the reactions of LMWOAs and heavy metals in soils are long-term, therefore, our experimental results can better reflect the actual situation. The results indicate that the ability of single LMWOAs (citric, oxalic, and malic acid) to affect the migration of Cr, Cd, and As followed the order citric acid > malic acid > oxalic acid at Sd d. The LMWOA-driven release of these heavy metals affected largely by the mobilization of Fe oxides and pH. The results of sequential extraction demonstrate that the LMWOAs induced the release of Pb, Cd, Cr, and As from soils, and advanced the reactivation of Pb, Cd, and As, but led to the immobilization of Cr in soil in this study. Results obtained in the study have implications for better understanding the long-term effect of plant root exudates on the mobilization of Cd, Cr, Pb, and As in coastal alkaline wetland soil environments.

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Disclosure statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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