

Sequential 'acid leaching–anion exchange–aerobic composting' process for recycle of municipal sludge

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ABSTRACT

Municipal sludge disposal and recycle has become a prominent research theme. In this study, a sequential process for integral treatment of municipal sludge was primarily presented, combining acid leaching, anion exchange and aerobic composting. The aim of the process was to remove chromium (Cr) from the sludge and reuse the sludge as manure. Firstly, Cr was removed from municipal sludge via the acid leaching process; the removal rate was up to 57.43%. Then, ion exchange resin was used to remove Cr from leachate; the removal rate reached 95%. Aluminum sheet was used to replace the Cr from eluent; the replacement rate was 63.3%. The aerobic composting process could be successfully warmed up to above 55 °C and lasted for 4 days; the seed germination index reached 68.3%. After the composting process, the residual Cr in sludge mainly existed at a more stable residual state and organic binding state. Overall, this novel sequential process serves as a potential high-efficiency, green, low-energy way for municipal sludge recycle.

Key words | aerobic composting, anion exchange, chromium, leaching, municipal sludge, recycling

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HIGHLIGHTS

- A 'leaching – ion exchange – composting' process for sludge recycle was studied.
- Cr content (544 mg/kg) met the standard of agricultural sludge after acid leaching.
- The replacement rate of Cr in eluent can reach 63.3% by Al replacing.
- Acid treated sludge reached maturity after aerobic composting for 12 days.

INTRODUCTION

Nowadays, municipal sludge treatment and disposal has become a prominent problem (Xing *et al.* 2011; Wang *et al.* 2017). In order to avoid secondary pollution, the sludge must be harmlessly treated and recycled (Wang *et al.* 2013a; Ko 2014; Wang 2017). Since sludge has high organic matter content and good fertilizer content, it can be used as a soil amendment and agricultural fertilizer. However, municipal sludge often contains high concentration of heavy metals, which limits the agricultural purposes of sludge (Kominko *et al.* 2019). To meet the agricultural standard, heavy metal removal is the key for sludge fertilization (Zhang *et al.* 2017).

At present, the heavy metals in sludge are usually passively removed rather than removed directly. However, through the

aerobic composting process, the chemical form of heavy metals in sludge may change and pollute the environment. Another main method is to use acid to oxidize heavy metals and change their state to dissolve them from sludge (Tyagi *et al.* 1991). For the acid removal process, the removal rate of some heavy metals by inorganic acids can reach more than 95% (Marchioretto *et al.* 2002). Nevertheless, the inorganic acids method is not applicable because of the large demand for acid. Meanwhile, the instrument is also easily corroded by strong acid (Igarashi *et al.* 2020). In contrast, the removal of heavy metals from sludge by organic acid is more promising, with a moderate pH value of 3–4 (Gunarathne *et al.* 2019). In addition, organic acids are degradable under aerobic or anaerobic conditions and

can be used for land use after composting, so this method has been widely used. The release of heavy metals was investigated using three organic acids (acetic, malic, and citric). The high rate of metals release by organic acids was explained through ligand-promoted mechanisms that enhance the release of metal ions from the sludge (Gunarathne *et al.* 2019). The acid oxidation of heavy metals only changes the state of heavy metals and dissolves them from sludge, but the subsequent acid wastewater treatment process has been seldom investigated. Meanwhile, it is also very important to examine the feasibility of the subsequent recovery of heavy metals after leaching and the reuse of sludge.

In this study, we carried out a study on a novel sequential process: 'acid leaching–anion exchange–aerobic composting' for Cr recovery and recycle of municipal sludge. This study mainly included three steps (Figure 1): (1) removal of Cr from sludge by citric acid leaching; then the obtained leachate was used to recover Cr, and acid treated sludge was used for aerobic composting; (2) Cr removal and recovery from leachate: removal of Cr from leachate by ion exchange resin, regeneration of ion exchange resin by two-step method; recovery of Cr by Al replacement; (3) aerobic compost and maturity assessment of acid treated sludge. The systematic research could provide a reference for municipal sludge recycling.

MATERIALS AND METHODS

Sludge source and basic properties determination

Concentrated sludge from a belt filter press of a municipal sewage treatment plant (Shenzhen, China) was used in this study. Total nitrogen (TN), total phosphorus (TP) and heavy metals (total As, Hg, Cr, Pb and Cd) in sludge were determined by the method described in the study by (Kominko *et al.* (2019)). The analysis of heavy metal existing forms was carried out by continuous extraction method (Naoum *et al.* 2001).

Separation of heavy metals from sludge by citric acid leaching

The test columns with inner diameter of 4 cm and length of 20 cm were used to load sludge. The column bottom was firstly fixed with nylon cloth, quartz sand (1.5 cm thick) was added, and then another layer of nylon cloth was laid on top. Afterwards, 90 g dry sludge (10 cm high) was loaded, and then a layer of nylon cloth with quartz sand (1.5 cm thick) was laid above it. After loading with sludge, the test column was fixed and soaked in deionized water until the sludge was wetted. The leaching dose was maintained at 500 mL/kg or

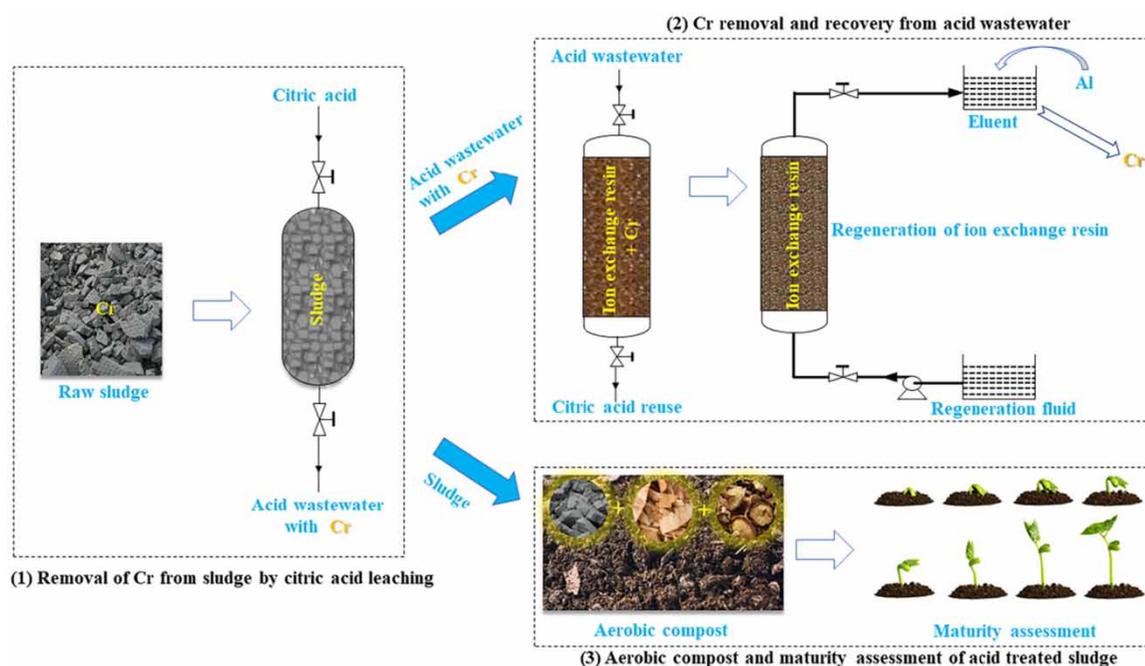


Figure 1 | Sequential acid leaching–anion exchange–aerobic composting process.

1,000 mL/kg, and the leaching speed was 0.5 mL/min. The content of total heavy metals in the leachate was determined. All the experiments were repeated three times.

Cr removal and recovery from leachate

Removal of Cr from leachate by ion exchange resin

Cr was separated from leachate by ion exchange resin; the process was divided into a static test and dynamic test.

- (1) Static test: 10 mL of leachate after citric acid leaching was mixed with 10 mL anion or cation exchange resin, and kept for 2 hours. The content of total Cr was determined after filtration by qualitative filter paper.
- (2) Dynamic test: 1 L of the leaching effluent was passed through 50 mL anion or cation exchange resin column at the flow rate of 5 mL/min. The content of total Cr in effluent was determined.

Regeneration of ion exchange resin by two-step method

After enriching with heavy metals, a two-step method was used to regenerate ion exchange resin. In the first step, 8% sodium hydroxide solution was used as regenerant. In the second step, a mixed solution of ethanol:sulfuric acid:water = 1:6.9:25 (weight ratio) was used as regenerant. Static and dynamic operation modes were also used in the regeneration process. The static method was to immerse ion exchange resins in the regeneration liquid. The dynamic method was that the regeneration liquid up-flowed slowly with a flow rate of 0.5 mL/min from the bottom to top of the resin column.

The amount of ethanol can be calculated according to the formula:

$$G = 0.66(MV - M'V')$$

G is the mass of ethanol (g), M is the content of Cr^{6+} in wastewater (g/L), V is the volume of exchange wastewater (L), M' is the content of Cr^{6+} in regenerated solution (g/L), V' is the volume of regenerated liquid (L) after regeneration with sodium hydroxide solution, and 0.66 is the conversion coefficient.

Recovery of Cr by Al replacement

In order to speed up the reaction, the oxide film on the surface of an Al sheet was removed by sandpaper, and then the Al sheet was cut into pieces ($2 \times 2 \text{ mm}^2$) and put into the heavy metal eluent for replacement reaction. The displaced

solution was sampled and analyzed every 8 hours, and the replacement reaction lasted for 3 days.

Aerobic composting of the acid treated sludge

The acid treated sludge was used as a composting matrix, sawdust and mushroom residue were used as composting fillers, and microbial fortifier CK-21 (DANO, Japan) was added into the composting fillers. The wet weight ratio of sludge:sawdust:mushroom residue was 35:5:7, C/N was 23.9, and the initial moisture content was 62.4%. The room temperature was 23–25 °C. A wooden board was laid on the bottom of the stack. Above the board, a layer of dry leaves about 15 cm thick was used as the stack bottom. All materials were evenly mixed and stacked. Each stack was about 1.0 m long, 0.5 m wide and 0.5 m high. The stack was covered with plastic cloth to keep warm and moist. The composting time was 15 days.

The evaluation of compost maturity can be divided into four categories: physical index, chemical index, biological index and spectral analysis method. In this study, five indexes – temperature, moisture content, pH, seed germination index (GI), and hygienic index – were selected to evaluate compost maturity and stability.

A compost sample (0.1 kg) was collected once a day for aerobic composting process analysis. For measuring sludge moisture content, pH and GI, the collected samples were mixed by multi-point mixing method, and then packed into a clean sealed bag. Some fresh samples were stored in a refrigerator at 4 °C. The moisture content was analyzed within 24 hours. The pathogenic bacteria were detected by MPN (most probable number) method. The other samples were laid in a tray and dried naturally. After grinding and sieving, the samples were extracted with distilled water at 1:10, and then analyzed by pH meter (Hanna, Italy). The seeds of water celery with high sensitivity and short germination time were selected for the germination test of sludge compost. The seed GI was calculated by the number of seeds germinated (seed germination) and seed root length.

$$\text{GI}(\%) = \frac{\text{seed germination} \times \text{root length of sample}}{\text{seed germination} \times \text{root length of control}} \times 100$$

This method has been used by the Italian government as a criterion for evaluating the maturity of organic waste and manure compost. The specific steps were as follows:

- After adjusting the moisture content of the compost sample to about 60%, the extract was obtained by pressure filtration.

- b. Seven seeds of water celery were placed in the extract solution and cultured in darkness at 27 °C for 24 hours. Fifteen repeats were set up.
- c. The germination and root growth of seeds were measured and expressed in percentage form. The GI is the percentage of the growth of the germination part and the root part.

RESULTS AND DISCUSSION

Properties of the municipal sludge

Nitrogen and phosphorus nutrients in sludge

Determining the quantity and composition of sewage sludge is a major component of designing sludge treatment units and their handling and disposal facilities (Najafzadeh & Zeinolabedini 2018; Zeinolabedini & Najafzadeh 2019). Municipal sludge contains a variety of plant nutrients, which is a necessary condition for agricultural utilization of sludge. Generally speaking, the higher the nutrient content of sludge, the more suitable it is for agricultural utilization (Kirchmann *et al.* 2017). In this study, TN and TP of the sludge were 22 g/kg and 12.9 g/kg, respectively. Compared with the statistical data of farm manure, the TN and TP of the sludge were higher than that of most farm manure (Khem *et al.* 2018), which indicated that the municipal sludge can be used for agricultural purposes.

Heavy metals content in sludge

In the raw sludge, the content of total Cr, As, Cd, Hg and Pb was 1,213, 7.0, 0.47, 94.4, 1.99 mg/kg, respectively. It can be seen that only the content of total Cr exceeded the agricultural sludge standard. For the heavy metal Cr in agricultural sludge, the limit in EU countries is no more than 1,000 mg/kg (86/278/EEC) (European Commission 1986). Germany has raised the standard on this basis, with the limit of 900 mg/kg (AbfKlaerV1992) (Germany 1992). The standards are somewhat stricter in China, requiring that the content of Cr in agricultural sludge cannot exceed 600 mg/kg (GB18918-2002 2002). Among the standards of heavy metals for agricultural sludge, there was no limit to As. The limit of Hg, Pb and Cd was 5, 300 and 5 mg/kg, respectively (86/278/EEC; AbfKlaerV1992; GB18918-2002), while the measured values of Hg, Pb and Cd were 0.47, 94.4 and 1.99 mg/kg, respectively. The values of Hg,

Pb and Cd were far below the standards, so it was needless to focus on them. Therefore, Cr was the only heavy metal in municipal sludge that exceeded the standard. And it was necessary to reduce concentration of Cr to below 600 mg/kg in sludge.

Removal of Cr from sludge by citric acid leaching

Citric acid can oxidize insoluble Cr^{3+} in sludge to more soluble Cr^{6+} and dissolve it from sludge (Neale *et al.* 1997). The sludge samples were first leached at a flow rate of 0.5 mL/min with the citric acid concentrations of 0.05, 0.1, 0.2, 0.4 and 0.8 mol/L. The removal rate of Cr is shown in Figure 2(a).

As shown in Figure 2(a), the removal rate of Cr increased with the increase of leaching dose and concentration. When the concentration of citric acid was 0.4 mol/L, the removal rate reached the maximum of 61.4%. The total residual Cr in sludge was 493.3 mg/kg.

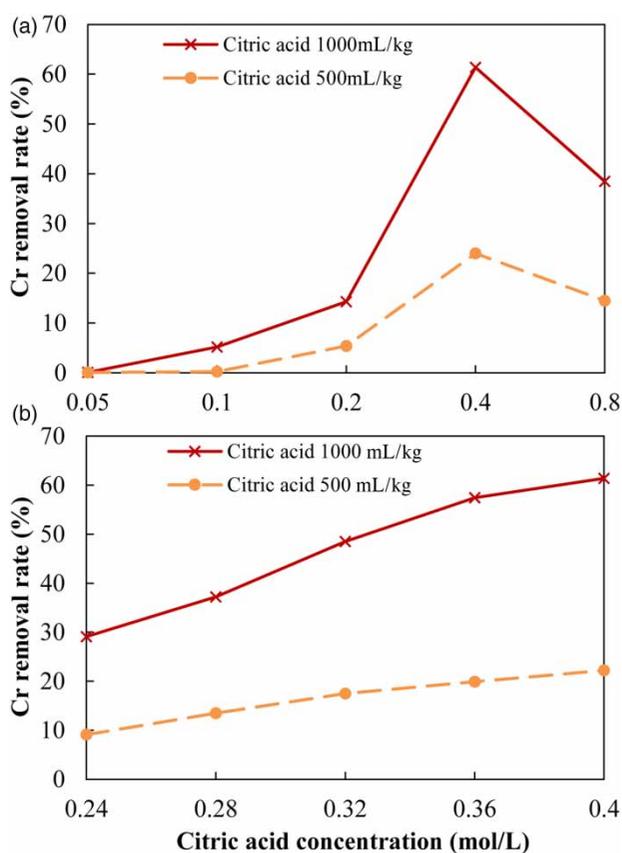


Figure 2 | Removal rate of Cr in sludge by citric acid leaching: (a) citric acid concentration 0.05–0.8 mol/L; (b) citric acid concentration 0.24–0.4 mol/L.

Then, in order to obtain more accurate citric acid concentration, 0.24, 0.28, 0.32, 0.36 and 0.4 mol/L citric acid was used for the above treatment processes. The results are shown in Figure 2(b). It can be seen that the removal rate of Cr with 0.36 mol/L citric acid could reach 57.43%, and the total residual Cr in sludge was 544.04 mg/kg, which also met the standards of GB18918-2002, Germany and other EU countries.

At a certain range of citric acid dosing (0.24–0.4 mol/L), increasing the concentration of acid for leaching can improve the removal rate of Cr. In order to save cost, 1,000 mL of 0.36 mol/L citric acid per kg of sludge was used for Cr leaching. The obtained leachate and acid treated sludge were used for the following study. And, for further study, models will be used, such as longitudinal dispersion coefficients, to obtain a more accurate Cr removal rate and to design more efficient reactors for leaching (Saberi-Movahed *et al.* 2020).

Removal and recovery of Cr from leachate

Removal of Cr from leachate by ion exchange resin

Ion exchange tests were carried out on leachate obtained in section 'Removal of Cr from sludge by citric acid leaching'. Both static and dynamic modes were operated for ion exchange. The results are shown in Figure 3. The analysis showed that anion exchange resin had a better removal rate on Cr in leachate with both static and dynamic exchange. Of these, the dynamic anion exchange resin achieved the optimum removal rate, which was over 95%.

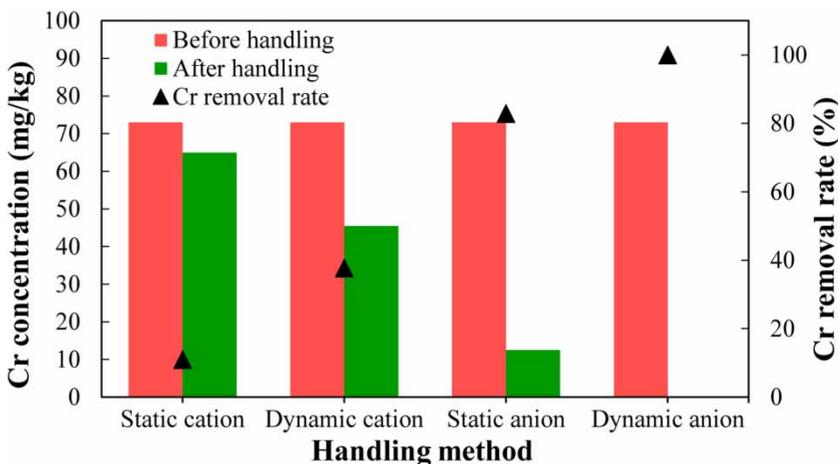


Figure 3 | Cr removal rate in citric acid wastewater by ion exchange resin. From left to right, four methods were used: static exchange of cation, dynamic exchange of cation, static exchange of anion and dynamic exchange of anion.

Regeneration of ion exchange resin by two-step elution method

The results of regeneration are shown in Figure 4. When the regenerated liquid volume was 50, 100 and 150 mL respectively, the dynamic elution rate was higher than the static elution rate. It can also be seen that when the regenerated liquid volume was 100 mL, the elution rate was obviously higher than that when the regenerated liquid volume was 50 mL, but if the regenerated liquid volume continued to increase, the elution rate did not increase significantly. Based on the above findings, the up-flow dynamic regeneration was selected and the volume of regenerated liquid

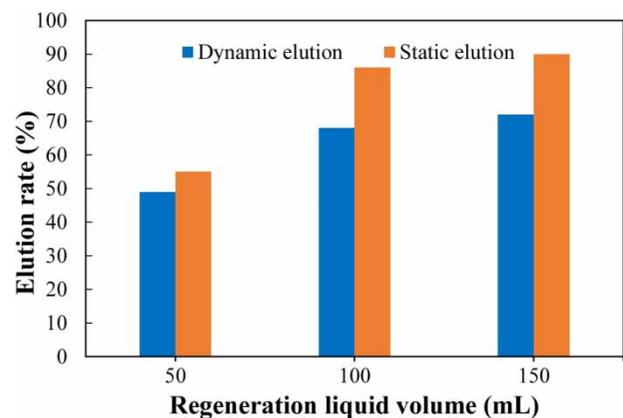


Figure 4 | The elution rate of ion exchange resin by two regeneration methods with different elution volume. Sodium hydroxide solution regeneration liquid: resin = 1–3:1 (volume ratio). Regenerated liquid volume was 50 mL, 100 mL and 150 mL, respectively. Static/dynamic elution modes were used. The regeneration liquid was up-flow with a flow rate of 0.5 mL/min.

was set as 100 mL. Therefore, when the regeneration solution:resin volume was 2:1, the up-flow dynamic elution rate can reach 86%.

Recovery of Cr by Al sheet replacement

Cr in the eluent can be replaced by zero-valent aluminum (Al) in acidic conditions at room temperature. The crushed Al sheets were directly added to the eluent of the elution process. Finally, the Cr containing mixture and chemical coagulants such as polyaluminum chloride can be obtained.

The Cr replacement rate under different replacement time is shown in Figure 5. With the replacement time increasing, the replacement rate increased gradually. When the replacement time was up to 72 hours, the replacement rate was 63.3%. On the one hand, it was related to the activity of metals. On the other hand, because of the low concentration of heavy metals in the regeneration solution, it was easy to be disturbed by other metal ions (such as high valence Al ions). If the dynamic replacement method and soft computing models are used to design the flow rate, higher replacement rate will be obtained (Najafzadeh & Zeinolabedini 2019).

Assessment of sludge maturity during aerobic composting

Temperature, moisture content, pH and GI in composting process

The change of temperature during the composting process is an important parameter for evaluating composting maturity. It is generally believed that if the temperature of the compost body is maintained for over 3 days at 55 °C, the pathogenic

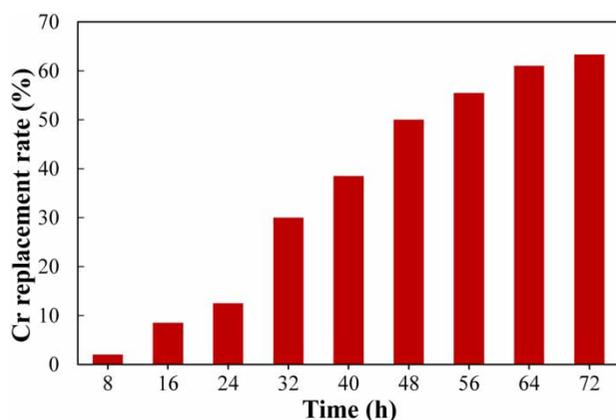


Figure 5 | The Cr replacement rates under different time by Al sheet replacing.

bacteria in the compost can be killed and the requirements of the compost hygiene index and compost maturity can be fulfilled (de Bertoldi 1998; de Bertoldi & Civilini 2006).

Figure 6(a) shows the composting temperature curve. After 1 day of composting, the temperature of the compost body rose rapidly to above 55 °C and was maintained for 4 days, accompanied by microbial metabolism and mass reproduction. After a high temperature period, the easily degradable organic matter has been basically exhausted, the microbial activity cannot produce enough heat, and the temperature of the compost body gradually decreased, approaching the environment temperature after 12 days. The change of temperature in the composting process generally includes three stages: heating (25–50 °C), high temperature persistence (above 50 °C) and cooling (35–50 °C).

Figure 6(b) shows the change curve of compost moisture content. Increasing composting temperature promoted water evaporation. The moisture content of compost decreased from 62.4% to 36.3%, with a decrease rate of 41.8%. The reduction was obvious and the sludge reduction effect was significant.

Figure 6(c) shows the change curve of compost pH. In the early stage of composting, production of organic acids by microbial decomposition of organic matter was greater than production of ammonia, and the pH decreased slowly to 6.56 on the fourth day. Then, organic matter decomposed by the strong action of microorganisms produced a large amount of NH₃. The pH increased gradually, and reached 7.24 at the 11th day, then decreased gradually. At the end of composting, pH was 7.03. Many researchers have proposed that the value of pH can be used as an index to evaluate the maturity of compost: in the initial stage of composting or fermentation, the value of pH is weak acid to neutral, generally 6.5–7.5; in the mature compost, it is generally weak alkaline, and the value of pH is about 8–9 (Jeong & Kim 2001; Liu *et al.* 2007). However, the pH value is also affected by the raw materials of compost. Thus, it can only be used as a reference index for composting maturity.

Figure 6(d) shows the variation of seed GI during composting. Generally speaking, when the seed GI of sludge is more than 50%, it indicates that the compost product has reached maturity (Xu *et al.* 2012; Wang *et al.* 2013b; Meng *et al.* 2018). In this experiment, the seed GI of sludge compost firstly decreased and then increased, and reached the lowest point on the fifth day. It was concluded that sludge composting at high temperature would produce a certain amount of NH₄⁺-N, organic acids, aldehydes and other substances, which might inhibit seed germination (Wang *et al.* 2018; Wang *et al.* 2019). With the decrease of composting temperature, the concentration of NH₄⁺-N and organic

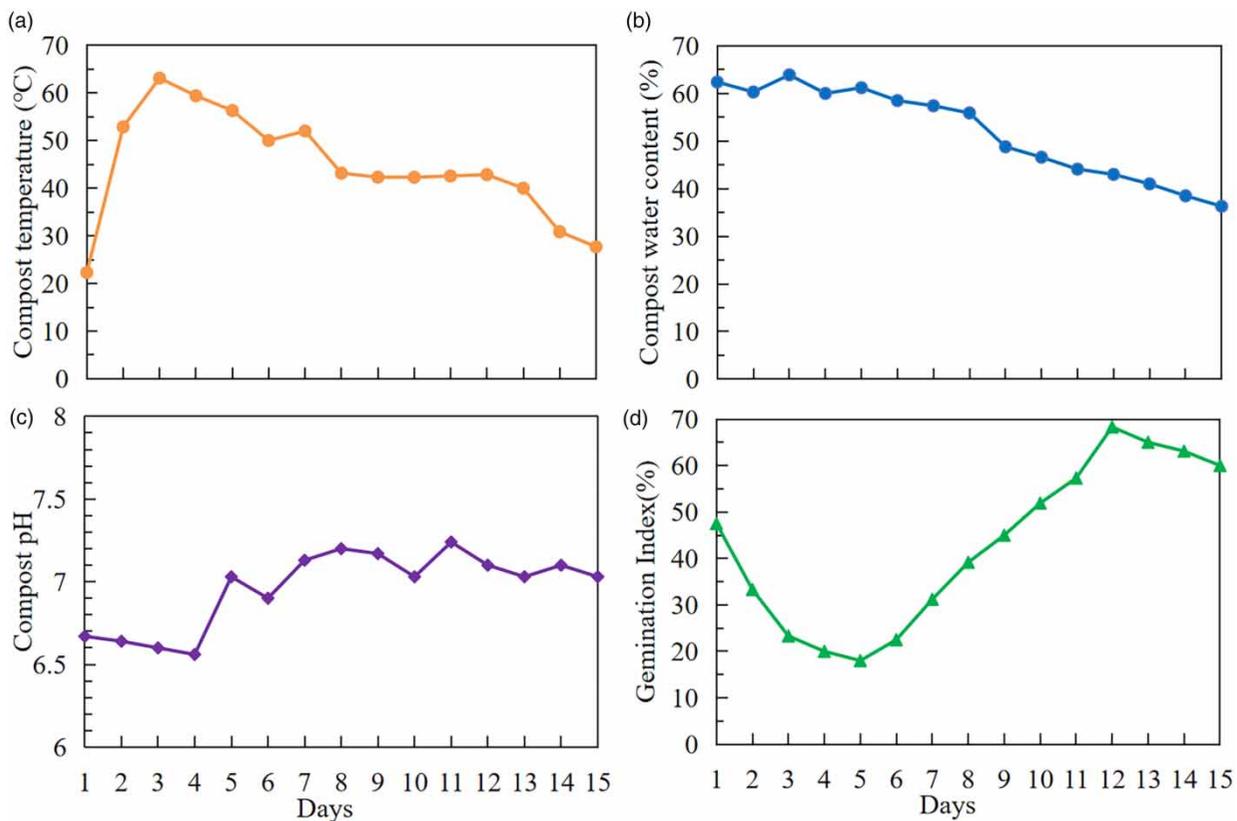


Figure 6 | The change of maturity indicators during sludge compost process: (a) composting temperature; (b) moisture content; (c) pH; (d) GI.

matter decreased, and GI increased gradually. GI increased to 68.3% on the 12th day, and remained above 50%. This indicated that sludge compost reached maturity after 12 days.

Hygienic indicators of composting

Salmonella and *Streptococcus entericus* are commonly used as indicators of safety for compost products. Health standards vary slightly in different countries and regions. Hsu and Lo suggested that 1 g compost contains less than 1 cell of *Salmonella* and 0.1–0.25 viral plaques, indicating that compost is stable and its application will not cause harm to human health and the surrounding environment (Hsu & Lo 2001). The Agricultural Standard of Grade A Sludge stipulated by the United States Environmental Protection Agency (US EPA) requires fecal coliform concentration <1,000 MPN/g, *Salmonella* concentration <3 MPN/4 g and *Ascaris* eggs <1 cell/4 g.

The hygienic indexes of compost products were tested. The results showed that the sterilizing rate of pathogenic bacteria in the composting process was 100%. All the hygienic indexes of the composting products could meet the

requirements of GB18918-2002 in China and grade A sludge agricultural standard stipulated by the US EPA, and could be used as manure.

Morphological analysis of Cr in compost products

After composting was matured, the speciation of heavy metal Cr in the sample was analyzed and compared (Table 1).

Studies showed that the order of stability of Cr in these states was as follows: residual state > organic matter binding state > carbonate binding state > iron-manganese oxide binding state > ion exchangeable state (Presley et al. 1980; Singh et al. 1999; Amir et al. 2005). As can be seen

Table 1 | Speciation of Cr before and after the composting

| | a | b | c | d | e | Total |
|-----------------------------|------|------|------|------|------|-------|
| Raw sludge (%) | 43.5 | 5.4 | 22.5 | 11.8 | 16.8 | 100 |
| Sludge after leaching (%) | 26.5 | 12.0 | 37.3 | 3.7 | 20.5 | 100 |
| Sludge after composting (%) | 50.8 | 32.0 | 12.1 | 1.8 | 3.4 | 100 |

a: residual state; b: organic matter binding state; c: carbonate binding state; d: iron-manganese oxide binding state; e: ion exchangeable state.

from Table 1, the exchangeable, carbonate binding and organic binding states in the sludge increased, while the proportion of residual states decreased, indicating that most of the Cr removed by citric acid existed in a stable form. After the treatment of citric acid leaching, the Cr in the sludge became unstable. However, after the aerobic composting process, Cr mainly existed in the residual state (50.8%) and organic-binding state (32.0%). The results showed that the stability of Cr was enhanced by aerobic composting. For the other heavy metals, citric acid increased the removal rate of Cd and Pb in sludge (Ma et al. 2020). In the sewage sludge compost under acid precipitation, the mobility of Cr, Cu and Pb decreased by 51–56% due to their retention by particulate organic matter, while the leaching of As, Cd and Ni was increased, as occurred in complexes (Qi et al. 2020). The increase of heavy metal stability demonstrated the feasibility of acid treated sludge compost for agricultural purpose.

CONCLUSIONS

A novel sequential citric acid leaching–anion exchange–aerobic composting process for Cr recovery and recycle of municipal sludge was conducted in this study. After citric acid leaching, the residual total Cr content in sludge was 544.04 mg/kg, which met the requirements of agricultural sludge. Meanwhile, the acid for leaching sludge can be recycled after anion exchange. And the anion exchange resin can be also reused after regeneration. The replacement rate of Cr in eluent can reach 63.3%. The results of the aerobic composting process for acid treated sludge showed that the compost products can be safe for agriculture use.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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