Citric acid and ethylene diamine tetra-acetic acid as effective washing agents to treat sewage sludge for agricultural reuse

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ARTICLE INFO

Article history:

Received 20 January 2015

Revised 12 July 2015

Accepted 14 July 2015

Available online 30 July 2015

Keywords:

Sewage sludge

Citric acid

Ethylene diamine tetra-acetic acid

Metal removal

Nutrients

FANP model

ABSTRACT

This paper presents the effects of different concentrations of citric acid (CA) and ethylene diamine tetra-acetic acid (EDTA) when used as additive reagents for the treatment of sewage sludge for agricultural use. Herein, both the retention of nutrients and removal of metals from the sewage sludge are examined. The average removal rate for the metals after treatment by CA decreased in the order $\text{Cu} > \text{Pb} > \text{Cd} > \text{Cr} > \text{Zn}$, while the rates after treatment by EDTA decreased in the order of $\text{Pb} > \text{Cu} > \text{Cr} > \text{Cd} > \text{Zn}$. After treatment with CA and EDTA, total nitrogen and total phosphorus concentrations in the sludge decreased, while the content of available nitrogen and Olsen-P increased. In addition, a multi-criteria analysis model-fuzzy analytic network process method (with 3 main factors and 12 assessment sub-factors) was adopted to evaluate the effectiveness of different treatment methods. The results showed that the optimal CA and EDTA concentrations for sewage sludge treatment were 0.60 and 0.125 mol/L, respectively.

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1. Introduction

Sewage sludge is a nutrient-rich material that normally contains high concentrations of carbon, nitrogen, phosphorous, organic matter, and mineral elements. Nutrients are essential for plant growth (Hanay et al., 2009; Singh and Sinha, 2004; Zhang et al., 2008), and the use of sewage sludge in agriculture to ameliorate the physicochemical deficiencies of degraded soil is a practice that is growing worldwide (Gupta and Sinha, 2006; Singh and Agrawal, 2007). Moreover, land application is considered one of the most economic and effective ways to dispose of sludge and supply plants with nutrients (Zhu et al., 2013). However, there are toxic metals in sewage sludge that necessitate restrictions on its use for certain land applications because of the potential for environmental and human health risks (Wang et al., 2012; Zhu et al., 2013). Therefore, there is need for sludge treatment methods that can reduce toxic metals while preserving nutrients, especially

if the sludge is to be reused for farmland. Over the past few decades, many methods for sewage sludge treatment and disposal have been developed. These include chemical treatments (Lo and Chen, 1990; Stylianou et al., 2007), thermal treatments (Zorpas et al., 2001), ion exchange methods (Dabrowski et al., 2004; de Villiers et al., 1995), chlorination methods (Fraissler et al., 2009), electrochemistry methods (Hanay et al., 2009), membrane separation methods (Chaudry et al., 1998), and bioleaching methods (Chen et al., 2005; Pathak et al., 2009). Chemical treatments have received extensive attention due to the simplicity of operation processes, short operation times, and high efficiency for metal removal (Deng et al., 2009). Various inorganic acids, organic acids, and strong complexing reagents have been proposed as effective additives to remove toxic metals from sludge or soil.

Citric acid (CA) and ethylene diamine tetra-acetic acid (EDTA) are the most widely used chemical additives because they have high efficiencies for metal extraction (Di Palma and Ferrantelli, 2005; Liu and Lin, 2013; Sun et al., 2001). Moreover, EDTA has been reported to have low aquatic toxicity and it does not bioaccumulate in living organisms throughout the food chain. However, its

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application may lead to nutrient deficiencies in the re-claimed sewage (Zhang et al., 2008). There are also other problems associated with the use of EDTA in remediation. For example, EDTA is recalcitrant in the environment and buildup of EDTA can lead to potential toxicity issues (Anupa et al., 2008; Peters, 1999). EDTA is biodegraded in soil under aerobic conditions at an estimated half-life of 120–300 days. Compared to EDTA, CA is more readily biodegradable (Römkens et al., 2002).

In this study, different concentrations of CA and EDTA were employed to treat sewage sludge to a level that would be sufficient for landfill requirements. As shown in previous studies, large doses and frequent applications of additives are typically needed to remove high concentrations of metals from sewage sludge (Deng et al., 2009; Zhu et al., 2009, 2013). However, such a large number of washings results in high processing costs and can even be detrimental to the environment because of the loss of nutrients (e.g., total nitrogen (TN), total phosphorus (TP), available-N, Olsen-P, and organic matter). Thus, it is necessary to determine optimal dosages and application frequencies for the additives to eliminate most of the toxic metals while simultaneously retaining most of the nutrients in the sludge.

The analytic network process (ANP) technique was proposed by Saaty (1996) to overcome the problem of interrelation among criteria or alternatives, and it has been successfully applied in many multi-criteria decision-making problems (Karsak et al., 2003; Yüksel and Dağdeviren, 2010). However, for problems such as the one posed by this study, i.e., those associated with incomplete information and subjective uncertainties, it can be difficult to quantify the precise ratio of weights for the different criteria used in each area. The concept of fuzzy sets has been incorporated into the ANP technique to deal with the problem of uncertainty. Hence, such a technique was applied in this study.

The primary objective of this research was to investigate the performance of different concentrations of CA and EDTA for treating sewage sludge to remove toxic metals and retain nutrients. In addition, the optimal conditions needed to maximize heavy-metal removal while minimizing nutrient loss were determined through the use of a fuzzy ANP (FANP) model.

2. Material and methods

2.1. Chelants

All the reagents used were of analytical grade. Additionally, the EDTA (ethylenediamine tetraacetic acid disodium salt dihydrate) and CA ($\text{H}_3\text{C}_6\text{H}_5\text{O}_7$, Sinopharm Chemical Reagent Beijing Co., Ltd) that were used in the laboratory experiments were of analytical grade. Deionized water was obtained from a Millipore Milli-Q system. All the standards, reagent solutions, and samples were stored in polyethylene containers pre-cleaned with 4.0 M HNO_3 and rinsed with deionized water.

2.2. Sampling of sewage sludge and soil

The sewage sludge used in the experiments was obtained from a two stage anaerobic sludge digester at a wastewater treatment plant (WWTP) located in Beijing, China. A total of 92.0% of the wastewater from the WWTP was domestic wastewater that was collected from a population of 800,000; 8.0% of the wastewater was industrial wastewater. The WWTP is operated as an extended aeration system. The sludge was air-dried, ground up, and passed through a 0.15 mm sieve. Sludge samples were then stored in polyethylene containers for further analyses and experiments.

The soil samples used in the experiments were collected from a fallow rice paddy in Yanglin City, Shanxi, China; these samples

were used to represent unpolluted soil. Adequate amounts of soil were collected from the topsoil (0–20.0 cm) and subsoil (20–40.0 cm) layers by use of an auger. The soil was also air-dried, ground up, and passed through a 0.15 mm sieve prior to being stored in polyethylene containers for further analyses and experiments.

The sewage sludge and soil samples that were collected were stored at 4 °C during transportation. After air drying the samples in the laboratory, physical and chemical characteristics of the samples were measured immediately. The physicochemical characteristics of the original sludge (OS) and soil are summarized in Table 1.

2.3. Sewage sludge and soil chemical analyses

Sludge and soil physical and chemical characteristics before and after washing treatments were respectively termed “original physicochemical characteristics” and “final physicochemical characteristics” in this study. Physicochemical characteristics measured included pH, relative humidity, total alkalinity, organic matter, TN, TP, available nitrogen, available P (Olsen-P), total suspended solids (TSS), volatile suspended solids (VSS), soluble chemical oxygen demand (SCOD), Cd, Pb, Cu, Zn, and Cr. These parameters were determined according to standard methods (APHA, 1998), except for available nitrogen, Olsen-P and metals. Available nitrogen was extracted from the soil at a soil solution ratio of 1:5 using 1.0 M KCl for 1 h. Afterward, Devarda alloy was added into the extracted solution to reduce the NO_3^- into NH_4^+ . The ammonia concentration was then determined by a titrimetric distillation method (EPA Method 350.2, U.S.) and the content of available nitrogen was obtained (Liu and Lin, 2013). Olsen-P was extracted using the Bray-1 method and quantified by the molybdenum blue colorimetric method (Olsen and Sommers, 1982). Soil and sludge samples were digested with a mixture of HCl/HNO_3 (Wu et al., 2004) and total concentrations of metals in soils and sludge were determined using an atomic absorption spectrometer (AAS, PERKIN ELMER Analyst 300).

A modified BCR-sequential extraction procedure (Aydin et al., 2013; Pan, 2009) was employed to study the partitioning of metals in the original sludge and treated sludge. This was done by separating the metals into fractions, namely, the exchangeable fraction, reducible fraction, oxidizable fraction, and residual fraction. After sequential washings, the sludge was air-dried and metal concentrations in the sludge samples were estimated for mass balance determination. Simultaneously, TN, TP, available-N, Olsen-P, and

Table 1
Physical and chemical characteristics of sludge and soil used in the experiment.

Parameters	Sewage sludge ^a	Soil ^a
pH	7.3 ± 0.1	6.8 ± 1.1
Relative humidity (%)	87.4 ± 0.30	78.8 ± 0.20
Total alkalinity (mg/kg)	81.2 ± 2.6	–
Organic matter (g/kg)	257.4 ± 12.8	10.6 ± 0.08
Total nitrogen (g/kg)	32.5 ± 0.47	1.1 ± 0.03
Total phosphorus (g/kg)	23.7 ± 0.21	5.1 ± 0.22
Available-N (mg/kg)	394.9 ± 8.9	116.7 ± 8.4
Olsen-P (mg/kg)	453.5 ± 44.4	26.4 ± 2.1
TSS (g/kg)	27.7 ± 0.12	–
VSS (g/kg)	20.8 ± 0.08	–
SCOD (mg/kg)	121.7 ± 18.3	–
Cd (mg/kg)	3.3 ± 0.58	0.16 ± 0.05
Pb (mg/kg)	54.7 ± 6.2	17.7 ± 2.3
Cu (mg/kg)	1735.4 ± 286.3	19.7 ± 0.69
Zn (mg/kg)	2110.3 ± 78.5	46.0 ± 3.3
Cr (mg/kg)	372.8 ± 23.6	9.3 ± 0.40

^a All these values are the means of four replicates. – no detect.

organic matter were measured in the air-dried sediment so that the retention of nutrients in the sludge could be evaluated.

2.4. Experimental approach

2.4.1. Target toxic metals and nutrients

CA and EDTA were used as additives in this investigation. Experiments were conducted to evaluate the performance of these additives in regards to both heavy-metal removal (i.e., Cu, Cd, Cr, Pb, and Zn) and the retention of nutrients (i.e., TN, TP, available-N, Olsen-P, and organic matter).

To explore the optimal dosage of CA and EDTA for the treatment of sludge, five different CA concentrations (0.20, 0.40, 0.60, 0.80, and 1.0 mol/L) and EDTA concentrations (0.025, 0.05, 0.10, 0.125, and 0.25 mol/L) were tested. Three grams of prepared sewage sludge (i.e., dry, ground, and sieved) were used in each experimental replicate. First, the sludge was oven dried at 103 °C for 24 h; then, it was added to 50.0 mL polyethylene containers containing 30.0 mL of the different concentrations of additives. The suspensions were mixed vigorously using a mechanical shaker with a constant speed of 150 rpm for 24 h at 25 °C. The suspensions were then centrifuged at 4000 rpm for 20 min and the toxic metal (Cu, Cd, Cr, Pb, and Zn) concentrations in the supernatants were determined by AAS.

2.4.2. Mixed sludge and soil

Sludge was mixed with prepared soil at ratios of 1:1, 1:2, and 1:4 at ambient temperature (25 ± 2 °C) and $60 \pm 5.0\%$ humidity within a ball mill. After mixing, the mixture (3 g) was dried at 103 °C for 24 h; then, it was added to 50.0 mL plastic containers containing 30.0 mL of the different concentrations of additives. The concentrations of toxic metals and nutrients of the mixed sludge and soil samples were quantified to ensure that the treated sludge could be used as a resource for soil reclamation.

2.5. Modeling methods

In this paper, fuzzy set theory was combined with the ANP technique into a FANP (fuzzy analytic network process) model to evaluate the performance of CA and EDTA for treating sewage sludge. The values of the linguistic variables assigned to the pairwise comparisons between the two criteria were converted into triangular fuzzy number (TFN) scores. Generally, a FANP model is developed in three steps that include (i) statement of the problem and development of the structure of the designed model, (ii) pairwise comparisons of the criteria used for the whole system to form the initial super-matrix, and (iii) formation of the weighted super-matrix.

In this study, five steps were used to develop the FANP model for the sewage sludge recycling and reuse treatment technology. The first step involved forming the network structure, which contained three factors and 12 sub-factors (see Fig. 1). In the second step, the pairwise comparison matrix was formed by assigning weights to the factors and sub-factors. The third step involved design of the evaluation standards and the development of an index by which to measure the performance of CA or EDTA treated sludge. In the fourth step, the super-matrix was established using the pairwise comparison matrix and the local weight vectors. In the fifth and final step, FANP data were collected and the rating scores for all indicators were calculated using the global indicator weights and linguistic values.

The methodology and development of the FANP model for evaluating sewage-sludge recycling-and-reuse treatment technology are discussed in detail in the [Supplementary Material](#).

Fig. 1. Proposed fuzzy analytic network process method for sewage sludge treatment methods.

3. Results and discussion

3.1. Sequential extraction

The results obtained after application of the sequential extraction scheme proposed by the BCR (Bureau Commune de Reference) for the original sewage sludge obtained from the WWTP are shown in Fig. 2. As Fig. 2 shows, most of the metals found in the sludge were in the exchangeable fraction. The percentages of Zn, Pb, Cd, Cr, and Cu in the original sludge bound to the extractable fraction were 57.5, 56.7, 48.1, 36.8, and 32.7%, respectively. In the original sludge, 37.6% of the Zn was found in the reducible fraction, whereas only 19.6, 14.4, 24.6, and 29.7% of the Cu, Cr, Cd, and Pb, respectively, were found in the reducible fraction. The oxidizable fraction contained 3.6, 1.5, 8.2, 4.7, and 1.1% of Zn, Cu, Cr, Cd, and Pb, respectively. Cu and Cr were obtained in the residual fraction at high levels of 46.2 and 40.6%, respectively.

3.2. Effects of CA and EDTA on the removal of metals

The results for the removal of metals from sludge with various concentrations of CA and EDTA are depicted in Fig. 3. The data clearly show that the removal efficiency for metals increased with increasing dosages of CA and EDTA. This proves that the addition of

Fig. 2. Metal fractionation of original sludge (OS).

Fig. 3. Removal of metals at different CA and EDTA concentrations. (a) Removal efficiencies for CA and (b) metals removed with CA; (c) removal efficiencies for EDTA and (d) metals removed with EDTA; OS means original sludge.

CA and EDTA as washing solutions is an effective way to promote toxic metal removal from sludge.

Washing sludge with CA was very effective for the removal of all tested toxic metals, and the highest average removal efficiency was observed for Zn (75.6%). The lowest average removal efficiency was observed for Cu (37.3%). This implies that CA can act as an effective washing agent for a variety of toxic metals. The average removal efficiencies for the toxic metals decreased in the order of Zn (75.6%) > Pb (69.9%) > Cd (58.7%) > Cr (45.0%) > Cu (37.7%) (Fig. 3a).

According to the results of Fig. 3a and b, the removal efficiencies for the toxic metals increased significantly with increasing CA dosages up to 0.60 mg/L. However, further increases in CA concentrations from 0.60 to 1.0 mol/L led to only a slight increase in the extraction of metals. This is probably because almost all the exchangeable and reducible fractions in the sludge were removed and the excess CA could not remove the oxidizable and residual fractions. In this study, dissolution of the metal–mineral bond followed by the dispersion of the pollutant metal in the washing liquid as an emulsion, complex, or suspension represent the processes that control the extraction of metals from contaminated sludge (Fuentes et al., 2008; Anupa et al., 2008).

The highest removal efficiencies for Zn, Pb, Cd, and Cr were 89.9 ± 5.1 , 86.5 ± 5.2 , 74.2 ± 4.8 , and $50.3 \pm 1.2\%$, respectively, and the corresponding metal removal amounts were 1897.8 ± 108.0 , 47.3 ± 2.8 , 2.5 ± 0.46 and 187.5 ± 4.5 mg/kg which were observed in 1.0 mol/L CA. In contrast, the highest Cu removal efficiency (49.1%) and removal amount (852.4 ± 22.4) were obtained at 0.80 mol/L CA.

Fig. 3c shows the removal rates for metals by EDTA. Five different concentrations of EDTA (0.025, 0.05, 0.10, 0.125, and 0.25 mol/L) were used in this research. The average removal rates

for the toxic metals decreased in the order of Pb (41.8%) > Zn (33.6%) > Cr (31.2%) > Cd (30.8%) > Cu (18.6%). The results show that when EDTA is used to wash sludge, only the exchangeable fraction can be removed. As shown in Fig. 3c and 3d, the highest removal efficiencies for Pb, Cd, and Cu were 47.8 ± 0.70 , 43.1 ± 1.0 , and $20.6 \pm 0.20\%$, respectively, with corresponding metal removal amounts of 26.1 ± 0.40 , 1.4 ± 0.03 , and 357.0 ± 3.1 mg/kg, respectively. The highest removal efficiencies for Zn and Cr were 39.1 ± 1.6 and $39.8 \pm 0.60\%$.

From the above analyses, we found that CA was associated with higher metal removal efficiencies than EDTA, and we can explain these results as follows. Metals extraction by CA and EDTA is based on the affinity of the organic ligand for the metals and the extent of complexation between polydentate organic ligands and heavy metals, which depends on the number and stability of metal-binding functional groups on the chelator. Thus, metals likely formed more stable complexes with the CA functional groups than with the EDTA functional groups and were therefore easily extracted in CA (Wang et al., 2015).

Two earlier research groups, namely those of Liu (Liu and Lin, 2013) and Anupa (Anupa et al., 2008), demonstrated that EDTA and CA could be used as chelating agents to remove metals from sludge or soil. In the most recent work (Liu and Lin, 2013), Cu removal efficiencies of 91.9 and 91.8% were obtained when 0.43 mol/L EDTA and 0.47 mol/L CA, respectively, were used to wash copper-contaminated soil. The earlier team (Anupa et al., 2008) observed the following metal removal amounts for sludge treated with EDTA over a period of five days: 77.0 mg/kg Cu, 42.4 mg/kg Pb, 23.6 mg/kg Cd, 6.4 mg/kg Cr, and 7220.0 mg/kg Zn. Furthermore, the results for treatment with CA decreased in the order of Cr (83.2 mg/kg) > Zn (77.0 mg/kg) > Pb (4.0 mg/kg) > Cu (2.5 mg/kg) > Cd (2.3 mg/kg). Though both CA

and EDTA were found to be efficient for metal removal from sludge, from the viewpoint of environmental protection, the use of high concentrations of EDTA (>0.125 mol/L) may be limited in practice. Additional insight into the economic effectiveness of using CA and EDTA for sludge treatment will be given in the later section on the FANP model results.

CA removes metals from sludge via two processes, namely acid-lysis and chelation. Multiple ligands can be observed in CA, and a stronger binding force exists between CA and heavy metal ions than for EDTA. The chelation and coordination reactions of EDTA are affected by the form of metals in the sludge, and the presence of more-available forms of metal can lead to higher removal efficiencies.

3.3. Effect of CA and EDTA on nutrients

The effects on nutrients following addition of various concentrations of CA and EDTA to the sludge were also tested. In these experiments, which were carried out under the same conditions as with the metal-removal experiments, it was anticipated that the use of CA and EDTA would reduce the nutrient concentrations in the sludge.

The nutrients were analyzed after treatment with CA and EDTA to determine the level of stabilization and quality of sludge (in terms of nutrient content) and to ensure that release of the treated sludge into the environment would be safe. The results (Figs. 3–5)

Fig. 4. Influence on TN and available-N for different (a) CA and (b) EDTA concentrations; OS means original sludge.

Fig. 5. Influence on TP and Olsen-P for different (a) CA and (b) EDTA concentrations; OS means original sludge.

show that the nutrients exhibited the same decreasing trends as the metals did during those experiments.

3.3.1. TN and available-N

The differences in nitrogen content (TN and available-N) in the sludge after treatment with the additives and as a percentage of their initial content are presented in Fig. 4.

Fig. 4 illustrates the effects of CA and EDTA on TN and available-N, and concentrations of 32.5 ± 0.47 g/kg for TN and 394.9 ± 8.9 mg/kg for available-N were obtained. This was about 1.2% of the available-N observed in the original sludge. During the experiments, changes in the TN concentrations of the sludge were not obvious when CA was added at low concentrations (0.20 and 0.40 mol/L). However, a decline of TN in the sludge was observed with increasing CA concentrations, and the lowest TN concentration (24.8 ± 1.3 g/kg) was obtained with the maximum concentration (1.0 mol/L) of CA. When the EDTA concentrations were varied from 0.025 to 0.50 mol/L, there was a corresponding decrease (27.5–30.0%) in the TN concentrations in the sludge. The lowest TN concentration (23.4 ± 0.84 g/kg) was observed when EDTA was added at 0.05 mol/L.

In most cases, knowledge of TN is not enough to characterize the capacity of sludge to supply nitrogen. This is because high TN does not mean that a high nitrogen supply will be available for plants. Therefore, available-N was also studied to provide a better characterization of the nitrogen content of the sludge. Fig. 4 shows that 0.20 mol/L CA had little influence on the removal of

available-N (1.5%). A decrease in available-N was observed with increasing CA concentrations. When the CA concentration reached 1.0 mol/L, the available-N of the sludge decreased to 201.3 ± 6.9 mg/kg (a drop of 49.0%). Compared with CA, EDTA showed different tendencies for the removal of available-N from sludge. At low concentrations (0.025 and 0.05 mol/L), EDTA showed a decreased tendency to reduce available-N. In contrast, at higher concentrations of EDTA (0.10, 0.125, and 0.25 mol/L), the available-N in sludge rose with increasing concentrations of the additive; the specific available-N concentrations in these treatments were 418.4 ± 2.5 , 432.3 ± 3.8 , and 477.8 ± 4.8 mg/kg, respectively, which correspond to increases of 5.9, 7.6, and 18.9%. These results are inconsistent with those from an earlier study (Liu and Lin, 2013), in which additives were employed to reclaim copper-contaminated soil, where the authors observed that the available-N content of the soil was slightly decreased after washing with EDTA or CA.

3.3.2. TP and Olsen-P

The effects of various CA and EDTA concentrations on the performance of TP and Olsen-P in sludge during the experiments are shown in Fig. 5.

Fig. 5 shows the amount of TP that was removed from the sludge after treatment with CA and EDTA. The TP and Olsen-P concentrations of the original sludge were 23.7 ± 0.21 g/kg and 453.5 ± 44.4 mg/kg (about 1.6% of TP), respectively. The TP content of the sludge clearly decreased when the CA concentration was increased from 0.20 to 0.60 mol/L (TP decreased to 6.5 ± 0.31 g/kg). This level was stable even when the CA concentration was further increased from 0.60 to 1.0 mol/L. It is noteworthy that the sludge-TP content did not fluctuate very much (22.4 ± 0.69 and 23.1 ± 0.50 g/kg) with corresponding EDTA concentrations of 0.025 and 0.25 mol/L.

Fig. 5 also shows that different CA and EDTA concentrations had little effect on the Olsen-P content. The concentrations of Olsen-P showed a slight increase when EDTA was used to treat the sludge, whereas a decreasing tendency was observed when CA was used to treat the sludge. The largest Olsen-P concentration of 485.4 ± 14.8 mg/kg, which is indicative of an increase of 7.0% compared with the original sludge, was observed when 0.25 mol/L EDTA was used to treat the sludge. However, Olsen-P in the sludge decreased from 453.5 ± 44.4 to 391.6 ± 24.2 , 397.6 ± 16.8 , 381.9 ± 12.7 , 379.6 ± 12.7 , and 384.3 ± 13.4 mg/kg with additions of CA at concentrations of 0.20, 0.40, 0.60, 0.80, and 1.0 mol/L, respectively. The tendency of change for phosphorus was the same as that reported earlier (Liu and Lin, 2013), where the authors observed that after treatment by EDTA or CA, the Olsen-P content increased to 1.3 to 5.1-times the original content. Wang et al. (2015) used CA along with an ultrasound process to extract metals from sludge and found that the total P was decreased from 34.9 to 18.3 g/kg after the extraction, while TN was decreased from 15.0 to 14.8 g/kg. They claimed that this sludge had good fertilization potential and could be used for soil amendments. From the above findings, it can be concluded that TP and Olsen-P will be increased in sludge treated by CA and EDTA.

The ratios of Olsen-P and TP increased when sludge was treated by CA and EDTA, especially when CA was employed to treat sludge, where the ratio of Olsen-P and TP increased from 1.6 to 5.9%. The increased ratio of Olsen-P and TP can possibly be interpreted by the fact that organic acids (such as CA) are strong chelators that can reduce phosphorus fixation and enhance the effectiveness of phosphorus by competition with anion adsorption reactions. However, many factors (e.g., pH, redox potential, leaching) can influence nitrogen and phosphorus removal from sludge. In this study, CA exhibited a higher TN and TP removal efficiency than EDTA due to the lower pH level (pH for CA treatments were

between 1.5 and 2.0, while pH for EDTA treatments were between 4.1 and 4.5). Washing under different pH conditions may have affected the dissolving and transforming processes for available-N and Olsen-P in the sludge. Moreover, a slight increase of available-N after washing with EDTA and decrease of Olsen-P after washing with CA were observed. There is one possible explanation for the increase of available-N and Olsen-P in the current study and that is as follows: mild acid washing might enhance the amount of available-N and Olsen-P by dissolving and transforming unavailable forms into available forms.

3.3.3. Organic matter

As can be seen from Fig. 6, comparisons of the original sludge to the sludge treated by CA show that the organic matter content increased from 257.4 ± 12.8 to 284.5 ± 13.6 g/kg (these are the average organic matter concentrations after five concentrations of CA were used to wash the sludge), and these values correspond to an increase between 9.3 and 12.0%. However, an opposite trend was observed with EDTA whereby the organic matter content decreased after treatment. The lowest organic matter content (200.3 ± 9.0 g/kg) was observed when the concentration of EDTA was 0.10 mol/L; thus, about 22.2% of the organic matter content was removed by EDTA.

This increase in organic matter content when CA was used to treat sludge can be attributed to a change in the structure of the sludge (Jakobsen et al., 2004; Hahladakis et al., 2014; Suèr et al., 2003; Wang et al., 2015). In previous studies, Wang et al. (2015) also observed increases after CA extraction; specifically, the organic matter content in the sludge from that study increased from 256.2 to 376.7 g/kg because of the residual citric acid, which may act as a nutrient source for fertilizers. Sun et al. (2011) demonstrated that CA could change the chemical properties at the sludge-surface interface and destroy organic-inorganic compounds thereby releasing a “bridge” of complex ions. When CA causes the sludge to form crumbs and clay, i.e., changing the sludge structure, organic matter is increased accordingly. In regards to

Fig. 6. Influence on organic matter for different (top) CA and (bottom) EDTA concentrations.

EDTA, the decrease of organic matter can be mainly attributed to leaching (Zhou and Wong, 2001).

3.4. Results for the application of the FANP model

It was necessary to determine the optimal CA and EDTA concentrations for metal removal and retention of nutrients to treat sewage sludge economically and in the most environmentally friendly way. Based on the [Supplementary Material](#), the FANP model was demonstrated to be a rather successful method for evaluating sewage-sludge-treatment technology.

Ten different additive-concentration conditions were explored using the FANP model (these are described in detail in the [Supplementary Material](#)). Take, for example, the case in which 0.60 mol/L CA was used to treat sludge. In this case, $B_{ad-CA(0.6)}$, $B_{mr-CA(0.6)}$, and $B_{ne-CA(0.6)}$ represent the numerical-scale values of the additive factor, metal-removal factor, and nutrient-elements factor using the fuzzy-ANP method, respectively. For $B_{ad-CA(0.6)}$, 89.0% of the respondents selected the 0.60 mol/L CA treatment as “fair” compared with nine other situations regarding the economic effectiveness of washing solutions. Moreover, 2.0, 8.0, and 1.0% of respondents deemed the treatment “good,” “poor,” or “very poor,” respectively. When considering the environmental effectiveness of 0.60 mol/L CA to treat sludge, 38.0 and 52.0% of respondents scored the treatment as “good” and “fair”, respectively, whereas the others selected “very good,” “poor,” and “very poor.” For $B_{mr-CA(0.6)}$ and $B_{ne-CA(0.6)}$, quantitative indicators can be described because Cr, Cu, Zn, Cd, and Pb removal efficiencies reached very high levels compared with the other situations. Thus, these degrees of membership were assigned 100% at the “very good” level. Consequently, the degrees of membership rated “good,” “fair,” “poor,” and “very poor” were all “0.”

The evaluation vectors of the sub-factors were computed by multiplying the local weight of the sub-factor with the scale-value to which it belonged. Computed global weights for sub-factors of 0.60 mol/L CA are shown as follows:

$$P_{ad-CA(0.6)} = w_{ws} \cdot B_{ws-CA(0.6)} = (0.459, 0.541) \cdot \begin{bmatrix} 0 & 0.02 & 0.89 & 0.08 & 0.01 \\ 0.01 & 0.38 & 0.52 & 0.08 & 0.01 \end{bmatrix} = (0.005, 0.215, 0.690, 0.080, 0.010) \quad (1)$$

$$P_{mr-CA(0.6)} = w_{mr} \cdot B_{mr-CA(0.6)} = (0.267, 0.184, 0.112, 0.067, 0.370) \cdot \begin{bmatrix} 1 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 \end{bmatrix} = (1, 0, 0, 0, 0) \quad (2)$$

$$P_{ne-CA(0.6)} = w_{ne} \cdot B_{ne-CA(0.6)} = (0.232, 0.177, 0.181, 0.143, 0.267) \cdot \begin{bmatrix} 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \\ 1 & 0 & 0 & 0 & 0 \end{bmatrix} = (0.267, 0.232, 0, 0.181, 0.320) \quad (3)$$

The evaluation vectors of the washing solution, metal removal, and nutrient elements were $P_{ad-CA(0.6)}$, $P_{mr-CA(0.6)}$, and $P_{ne-CA(0.6)}$, respectively. According to the calculations made, significant differences were observed in the results obtained for the sub-factors when interdependent priorities of the criteria and dependencies

were not taken into account. For example, the results changed from 0.232 to 0.267, from 0.177 to 0.232, from 0.181 to “0,” from 0.143 to 0.181, and from 0.267 to 0.320 for the sub-factor global-weight values describing the influence of TN, TP, available-N, Olsen-P, and organic matter, respectively.

$$P_{CA(0.6)} = w \cdot B_{CA(0.6)} = (0.211, 0.489, 0.301) \cdot \begin{bmatrix} 0.005 & 0.215 & 0.690 & 0.080 & 0.010 \\ 1 & 0 & 0 & 0 & 0 \\ 0.267 & 0.232 & 0 & 0.181 & 0.320 \end{bmatrix} = (0.571, 0.115, 0.146, 0.071, 0.098) \quad (4)$$

In the next stage, the evaluation vector of the goal $P_{CA(0.6)}$ was obtained by multiplying the global weights of factors (0.211, 0.489, 0.301) with the evaluation vector of sub-factor $B_{CA(0.6)}$. According to these calculations, the “very good,” “good,” “fair,” “poor,” and “very poor” scales for 0.60 mol/L CA were determined to be 57.1, 11.5, 14.6, 7.1, and 9.8%, respectively.

The last step involved the calculation of the comprehensive score of the goal by multiplying the evaluation vector and evaluation class vector (1.0, 0.75, 0.50, 0.25, “0”) as follows:

$$S_{CA(0.6)} = P_{CA(0.6)} \cdot s' = (0.571, 0.115, 0.146, 0.071, 0.098) \cdot \begin{pmatrix} 1 \\ 0.75 \\ 0.5 \\ 0.25 \\ 0 \end{pmatrix} = 0.748 \quad (5)$$

The comprehensive scores of the left nine situations were calculated the same as for the above methods, and the evaluation vectors and comprehensive scores are listed in [Table 2](#). According to the calculations, these values suggest that CA has a better comprehensive score for performance when using the FANP model to evaluate the effectiveness of the different washing solutions (CA and EDTA) for treating sludge. As [Table 2](#) shows, the largest comprehensive score (0.748) was obtained when 0.60 mol/L CA was employed to treat sludge. In regards to EDTA, the largest comprehensive score (0.419) was obtained when 0.125 mol/L EDTA was employed to treat sludge.

3.5. Results for mixed sludge and soil

Based on the comprehensive scores provided by the FANP model, the selected optimum dosages of CA and EDTA under the experimental conditions tested were determined to be 0.60 mol/L and 0.125 mol/L, respectively. To evaluate the treated sludge for agricultural use more specifically, an experiment was conducted using a mixture of sludge and soil. The results are presented below.

Table 2
Degree of relative impact for different additive reagents.

Concentration(mol/l)	Evaluation vector	Score
<i>(a) Degree of relative impact for CA</i>		
0.20	(0.339, 0.205, 0.055, 0.123, 0.279)	0.551
0.40	(0.261, 0.360, 0.229, 0.043, 0.108)	0.656
0.60	(0.571, 0.115, 0.146, 0.071, 0.098)	0.748
0.80	(0.569, 0.002, 0.126, 0.138, 0.157)	0.668
1.0	(0.569, 0.043, 0.005, 0.016, 0.371)	0.608
<i>(b) Degree of relative impact for EDTA</i>		
0.025	(0.242, 0.115, 0.152, 0.000, 0.489)	0.405
0.05	(0.261, 0.090, 0.011, 0.211, 0.428)	0.378
0.10	(0.172, 0.173, 0.147, 0.033, 0.476)	0.384
0.125	(0.153, 0.073, 0.275, 0.296, 0.205)	0.419
0.25	(0.281, 0.000, 0.006, 0.176, 0.538)	0.328

Table 3

Concentration of heavy metals and nutrients in sludge treated with 0.60 mol/L CA at different sludge/soil ratios.

Sludge: soil	Metals (mg/kg)					TN (g/kg)	TP (g/kg)	Available-N (mg/kg)	Olsen-P (mg/kg)	Organic matters (g/kg)
	Cd	Pb	Cu	Zn	Cr					
A-grade	<3.0	<300.0	<500.0	<1500.0	<500.0	–	–	–	–	–
Sludge	1.0	9.1	326.5	1116.1	202.2	29.6	6.5	264.0	409.1	300.7
1:1	0.83	12.1	157.2	627.6	93.7	13.8	5.7	236.8	288.6	143.2
1:2	0.52	12.7	101.4	355.8	37.3	13.0	5.2	198.3	234.7	94.6
1:4	0.34	15.7	78.9	122.3	32.0	5.9	5.4	153.8	184.6	66.0
Soil	0.16	17.7	19.7	46.0	9.3	1.1	5.1	116.7	26.4	10.6

Table 4

Concentration of heavy metals and nutrients in sludge treated with 0.125 mol/L EDTA at different sludge/soil ratios.

Sludge: soil	Metals (mg/kg)					TN (g/kg)	TP (g/kg)	Available-N (mg/kg)	Olsen-P (mg/kg)	Organic matter (g/kg)
	Cd	Pb	Cu	Zn	Cr					
B-grade	<15.0	<1000.0	<1500.0	<3000.0	<1000.0	–	–	–	–	–
Sludge	1.9	28.6	1060.3	1676.2	232.1	23.2	22.0	432.3	498.0	221.9
1:1	1.2	26.2	553.2	875.7	126.5	11.7	12.8	414.6	468.7	105.9
1:2	0.79	19.4	310.8	429.5	57.9	9.4	8.6	342.9	313.1	66.6
1:4	0.62	21.1	129.4	376.8	50.2	4.9	7.3	283.4	198.5	39.7
Soil	0.16	17.7	19.7	46.0	9.3	1.1	5.1	116.7	26.4	10.6

3.5.1. Results for the mixture of CA-treated sludge and soil

The concentrations of toxic metals and nutrients in the sludge confirmed that it would be possible to use treated sewage sludge in agriculture. In fact, after treatment with 0.60 mol/L CA, the sludge concentrations of the toxic metals Cr, Cu, Zn, Cd, and Pb decreased to 202.2, 326.5, 1116.1, 1.0, and 9.1 mg/kg, respectively (Table 4). As for the nutrients TN, TP, available-N, Olsen-P, and organic matter, the concentrations were 29.6 g/kg, 6.5 g/kg, 264.0 mg/kg, 409.1 mg/kg, and 300.7 g/kg, respectively. These were higher than the nutrients concentrations in the soil. The residual contents of metals and nutrients in the treated sludge meet China's requirements outlined in the Disposal of Sludge from Municipal Wastewater Treatment Plants-Control Standards for Agricultural Use (CJ/T 309-2009) for A-grade treated sludge.

The data in Table 3 demonstrate that all of the toxic metals except for Pb decreased in the mixed sludge–soil sample after adjustment for the sludge/soil ratio. The optimal ratio of mixed CA-treated sludge and soil for agricultural reuse was determined to be 1:2; at this ratio, the concentrations of Cd, Pb, Cu, Zn, and Cr in the mixed sample were 0.52, 12.7, 101.4, 355.8, and 7.3 mg/kg, respectively. The content of TN, TP, available-N, Olsen-P, and organic matter were 13.0 g/kg, 5.2 g/kg, 198.3 mg/kg, 234.7 mg/kg, and 94.6 g/kg, respectively.

3.5.2. Results for the mixture of EDTA-treated sludge and soil

The metal and nutrient concentrations of sludge treated with 0.125 mol/L EDTA are depicted in Section 3.2 and Table 4. The concentrations of Cr, Cd, and Pb (232.1, 1.9, and 28.6 mg/kg, respectively) did not exceed the regulatory limits (for A-grade sludge, as described in CJ/T 309-2009) for agricultural amendments, which are 3.0, 500.0, and 300.0 mg/kg for Cr, Cd, and Pb, respectively. However, the concentrations of Cu and Zn in the treated sludge were 1060.5 and 1676.2 mg/kg, respectively, and these concentrations were higher than the A-grade sludge requirements. The concentrations of TN, TP, available-N, Olsen-P, and organic matter were 23.2 g/kg, 22.0 g/kg, 432.3 mg/kg, 498.0 mg/kg, and 221.9 g/kg, respectively. These results show that after treatment with 0.125 mol/L EDTA, the sludge could be used as B-grade sludge according to the official guidelines of China.

Table 4 shows that lower concentrations of toxic metals could be achieved by mixing more soil into the mixed sample, and the

optimal ratio of sludge mixed soil was 1:2 (at which the concentrations of Cd, Pb, Cu, Zn, and Cr in the mixed sample were 0.79, 19.4, 310.8, 429.5, and 57.9 mg/kg, respectively). The concentrations of TN, TP, available-N, Olsen-P, and organic matter were 9.4 g/kg, 8.6 g/kg, 342.9 mg/kg, 313.1 mg/kg, and 66.6 g/kg, respectively.

4. Conclusions

Sewage sludge treated with CA and EDTA successfully met the Chinese guidelines for land application, so the method used in this study represents a reliable and economic way to solve the sludge problem. Both CA and EDTA showed good performance for removing metals from the sewage sludge. Although the total contents of nitrogen and phosphorus decreased when sewage sludge was treated with CA and EDTA, the contents of Olsen-P and available-N, which are appropriate forms for plant use, actually increased. Therefore, there should be no concerns about nutrient loss as a result of the washing agent treatment.

In addition, according to our new FANP model, which was developed to determine the optimal treatment conditions, the best outcomes for sludge treatment can be achieved with CA and EDTA concentrations of 0.60 and 0.125 mol/L, respectively.

Acknowledgements

This research was supported by the Scientific Research Common Program of the Beijing Municipal Commission of Education (KM201210016011) and the Beijing Academic Innovation Group in Urban Storm Water System and Water Environmental Eco-Technologies (PHR201106124). Additionally, this study was supported by grant Z141109004414087.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.wasman.2015.07.021>.

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