Research article

Heavy metal removal from sludge with organic chelators: Comparative study of N, N-bis(carboxymethyl) glutamic acid and citric acid

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ABSTRACT

The applicability and performance of a new generation of biodegradable chelator, N, N-Bis(carboxymethyl) glutamic acid (GLDA), for extracting heavy metals from sewage sludge was carried out and compared with citric acid (CA). Targeted metals included Cd, Co, Cu, Zn, Ni and Cr, and their contents in the raw sludge were 63.1, 73.4, 1103.2, 2060.3, 483.9 and 604.1 mg kg^{-1} (dry sludge basis), respectively. Metals were divided into six fractions including water soluble, exchangeable, carbonates bound, Fe–Mn bound, organic matters bound and residual fraction via chemical fractionation. Washing results showed that in general GLDA exhibited better performance compared with CA, with removal efficiency of 83.9, 87.3, 81.2, 85.6, 89.3 and 90.2% for Cd, Co, Cu, Zn, Ni and Cr, respectively at equilibrium pH = 3.3. Residual metals were better stabilized in the GLAD-washed sludge than in the CA-washed sludge, and were mostly tightly bonded to the residual fraction. Furthermore, CA promoted phosphorus (P) release while GLDA had an opposite effect and tended to retain P within sludge, which could be beneficial for further application in agricultural use. Findings from this study suggested that GLDA could be a potential replacement for refractory and less environmentally-friendly chelators in the extraction of metals from sludge.

1. Introduction

It is noteworthy that the use of sludge in agriculture has many advantages, which include providing recyclable nutrimental elements including phosphorus, nitrogen and potassium for plant growth, and organic matters which empower the soil with numerous numbers of beneficial organisms, reduce the need for synthetic fertilizers and pesticides, and improve the physical and biological properties of the soil (Wang, 1997; Kvasauskas and Baltrenas, 2009; Pend et al., 2011). Despite its many advantages, it is now widely accepted that the use of sewage sludge in agriculture as soil fertilizer or for land application has a potential health concern because of the plethora of pollutants that it contains, such

as heavy metals. Unlike organic compounds that can be biodegraded with time, heavy metals are not biodegradable, which makes them a potential threat to the environment and human heath (Bailey et al., 1999; Hong et al., 2002; Wuana et al., 2010). Heavy metals can enter food chain through the uptake by plants from soil, and finally end up in human. High concentration of heavy metals in sludge is therefore one of the major obstacles limiting its agricultural application. Therefore, to achieve a more environmentally friendly sludge, it is important to consider the post treatment of sludge by stabilizing or lowering the contents of heavy metals before its land application.

Chemical leaching is one of the common and widely used methods for soil and sludge's heavy metals removal. Generally, heavy metal-contaminated soil or sludge is treated with strong inorganic acids such as HCl, HNO$_3$, H$_2$SO$_4$, H$_3$PO$_4$ (Stylianou et al., 2007) and chelating reagents such as nitrilotriacetic acid (NTA)

(Babel and del Mundo Dacera, 2006), sodium tripolyphosphates (STPP) and ethylenediaminetetraacetic acid (EDTA) (Yu and Klarup, 1994; Babel and del Mundo Dacera, 2006; Di Palma et al., 2007). The application of above-mentioned strong acids and chelating agents gives very good recovery of various metals. However, on one hand, their negative impacts on soil or sludge structure might affect the fertility and plants productivity (Cline and Reed, 1995; Lee and Marshall, 2002); on the other hand, specifically, NTA and EDTA exhibit refractory behavior towards biodegradation and can pose a secondary pollution via leaching to the groundwater (Nowack, 2002). Therefore, replacement of such chelators with new environmentally-friendly ones is highly recommended. Organic acids and new generation of chelating agents have been increasingly suggested as alternatives to above-mentioned strong acids and chelating agents. Low molecular weight organic acids such as acetic acid, oxalic acid (Zaleckas et al., 2013) and citric acid (CA) (Di Palma et al., 2007; Zaleckas et al., 2013) have been reported to be promising alternatives for replacing refractory chelating agents. Recently, novel readily biodegradable chelators such as N, N-bis(-carboxymethyl) glutamic acid (GLDA) has been suggested as an alternative for heavy metal recovering from soil due to its exceptional chelating capacity towards different divalent metal ions (Kołodyńska, 2011). It also possesses excellent biodegradability (Seetz and Stanitzek, 2008). According to the OECD 301D test (Schneider et al., 1999), more than 60% of GLDA can be degraded within 28 days. Moreover, Borén et al. (2009) reported that it has the lowest 'eco-footprint' characteristics in comparison to EDTA and STPP.

To our knowledge, comparative study of the use of GLDA for sludge's heavy metal removal with other chelator(s) is not yet documented. Therefore, this study aimed at first, to investigate the applicability of GLDA for heavy metals extraction from sewage sludge and compare its performance with CA and secondly, to investigate the impact of the washing procedure on the changes of metal stabilization via chemical fractionation. Finally, we examined the influence of both organic chelators on the mobilization of macronutrient, such as phosphorus in the sludge. Our results will provide insights on GLDA potential as an environmentally-friendly chelator for heavy metal removal from sludge.

2. Materials and methods

2.1. Sludge and chemical reagents

Dewatered sludge used in this study was collected from a local municipal wastewater treatment plant (Xiamen, China), and was at first air dried before further dried in an oven at 102 ± 3 °C. Dry sludge was grounded in a mortar and sieved through a mesh size <0.15 mm in diameter and stored in the plastic bag. Chelating agents used included CA and GLDA. CA monohydrate (99.9%) was purchased from Sinopharm Chemical Reagent Co. Ltd. and GLDA (MW = 351.13 g mol^{-1}, solid content (40%) and density = 1.35 g cm^{-3}) was purchased from TCI Development Co. Ltd. (Shanghai, China).

2.2. Chemical washing and precipitation

For heavy metal solubilization, the previously prepared sludge was subjected to chemical washing using CA and GLDA. Washing experiments were carried out at room temperature (28 °C) in 120 mL glass bottles pre-washed with acidic solution and rinsed with Milli-Q water. Each extracting solutions were prepared to the concentration of 100 mM by dissolving the corresponding reagent salt into the Milli-Q water, and solutions of different pH values (1—6) were further prepared. Values of pH were adjusted by H_2SO_4 or NaOH. Sludge and extracting solutions were then mixed

at the ratio of 1:10 (w:v). Controls with Milli-Q water were also set up. Mixtures were hand-shaken for about 2 min and then mechanically shaken at 250 rpm for 12 h (for the effect of pH) and 24 h (for effect of contact time). Two point five milliliters of samples from each bottle were collected at the beginning and end of the process for determination of the optimal pH. To investigate the effect of contact time, experiments were conducted at the optimal pH. Two point five milliliters of samples from each bottle were periodically collected at 0, 3, 6, 12 and 24 h and were filtered through 0.45 μm Millipore filter, diluted 20 times and stored under 4 °C prior to metal analysis. Each experiment was conducted in duplicate and the average concentrations were used in this study. At the end of the process, solubilized metals were precipitated with sodium hydroxide solution (10^{-2} M) and the recovery was calculated.

2.3. Analytical methods

2.3.1. Physicochemical characterization

Physicochemical characterization of raw sludge including pH, moisture content (Mc), electrical conductivity (EC), total organic matter (OM) and total phosphorus (TP) were determined and results are presented in the Table S1 in Supporting Information (SI). Mc was determined by drying sludge samples in the oven at 102 ± 3 °C. EC and pH were determined through the supernatant of dissolved sludge in the Milli-Q water under the ratio 1:10 (w:v) after shaken for 30 min and centrifuged at 5000 rpm for 15 min. OM was determined by weight loss by ignition at 600 °C for 6 h. TP was determined by aqua regia digestion according to ISO 11464, EN 12880 and quantification was made by Flow Injection Analyzer (Lachat QC8500, USA). Heavy metals including cadmium (Cd), Cobalt (Co), copper (Cu), zinc (Zn), nickel (Ni) and chromium (Cr) contents and species distribution were also determined.

2.3.2. Metal fractionation

Chemical fractionation of targeted heavy metals before and after the washing process was carried out according to Tessier et al. (1979) with slight modification (Aikpokpodion et al., 2013) as shown in Table S2 in SI. Metals were divided into 6 different fractions as follows: Water soluble (F1), exchangeable (F2), carbonates bound (F3), iron and manganese-bound (F4), organic matters bound (F5) and residual (F6). The procedure from Chen and Ma (2001) was used for extracting residual fraction of metals, and the same procedure was used to perform parallel total digestion of the same amount of sludge and sediment reference material (GBW07309, General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China) in order to calculate the recovery efficiency of the sequential extraction procedures.

2.3.3. Stock and standard solution preparation and metal analysis

Stock solutions (1000 mg L^{-1}) of the targeted heavy metals were prepared by dissolving a well weighted amount of their corresponding salt in Milli-Q water. Dissolved salts included $(CdCl_2)_2 \cdot 5H_2O$, $CoCl_2 \cdot 6H_2O$, $CuCl_2 \cdot 2H_2O$, $ZnCl_2$, $NiCl_2 \cdot 6H_2O$ and $K_2Cr_2O_7$. Stock solutions were stored at 4 °C and working standard solutions (0, 2, 4, 10, 25, 50, 75 and 100 mg L^{-1}) were prepared by appropriate dilution with Milli-Q water. Metals in original sludge and all collected samples were analyzed using Inductively Coupled Plasma Optical Emission Spectrometry (Perkin Elmer Optima 7000 DV, USA).

3. Results and discussion

3.1. Metal contents and fractionation in the original sludge

Metal contents in the original sludge were 63.1, 73.4, 1103.2, 2060.3, 483.9 and 604.1 mg kg^{-1} (dry sludge basis), for Cd, Co, Cu. Zn, Ni and Cr, respectively. According to Chinese National Environmental Standard quality for usable sludge in agriculture (National Standard GB 4284, China), heavy metal concentration in the sludge which could be used in agriculture is fixed as follows: 5, 600, 800, 100 and 2000 mg kg^{-1} for acidic soil (pH $\leq$ 6.5) and 20, 1000, 1500, 200 and 3000 mg kg^{-1} for alkali soil (pH $\geq$ 6.5) regarding Cd, Cr, Cu Ni and Zn, respectively. It can thus be stated that metal contents of the original sludge are not in agreement with the regulation for the agricultural usage. When comparing with other countries, the metal contents also exceed the allowed limits in European countries such as Netherlands, Sweden and Denmark (0.8–8, 75–600, 75–100, 30–50 and 300–800 mg kg^{-1} (dry sludge basis), respectively) (Salado et al., 2008; Olofsson et al., 2012). Zn and Cu due to their high concentrations are the major metals in the original sludge. This could be because Zn and Cu are commonly used as macronutrient elements in foodstuff additives to promote growth rate, which consequently resulted in their accumulation in sludge (Xiong et al., 2010). In addition, other sources such as galvanized materials and car washes were reported for Zn and leaching from the new plumbing systems was reported as the source for Cu (Sörme and Lagerkvist, 2002; Rule et al., 2006).

It would be important to determine the different forms or states in which each metal predominates in the sludge (through sequential fractionation) for batter evaluating the agricultural use of the sludge. Details of chemical fractionation of metals are shown in Table 1. It can be noticed that metal distribution in the sludge varies from one fraction to another depending on the type of metal. Target metals were mostly found in the form of organic matters (F5), carbonates (F3), subsequently Fe–Mn oxides (F4) and residual (F6) fraction. Unlike other metals, Cd was predominantly found in F3 (48.8%) as previously reported by Gao et al. (2013), while Cu, Zn, Ni and Cr predominated in the form of F5 (35.1, 44.4, 51.3 and 49.4%, respectively) followed by F3 (27.1, 20.7 and 23.6% for Cu, Zn and Ni) and F2 (16.8% for Cr). All metals exhibited low concentrations in water soluble (F1) and exchangeable (F2) forms except that a larger amount of Cr was bonded to F2 (16.8%). About 7–25% of total contents (depending on the metal) were fixed to the residual fraction (F6). According to Achiba et al. (2010), the sum (F1 + F2 + F3) is the minimum bioavailable proportion of a given metal in the soil, which can be easily released to the environment and uptake by plants. It is highly sensitive to the environmental conditions and more available to biota (Labanowski et al., 2008). In the current work, it represents about 50, 30, 35, 25, 30 and 32.5% for Cd, Co, Cu, Zn, Ni and Cr, respectively, while the sum (F1 + F2 + F3 + F4 + F5) for each metal ranged from 75% (for Cu) to 93% (for Cr). As a consequence, metals were not well stabilized within the sludge and might be easily mobilized and exhibit negative impacts on the environment and ecosystem once the sludge is poured or spread into the environment, especially Cd, which was remarkably found in F3 (>45%).

3.2. Effect of pH on washing efficiency

Results of chemical washing of the sludge are presented in Fig. 1. Based on the data, target metals exhibited various behaviors towards chelating agents depending on the equilibrium pH of the mixture (pHe). In the absence of chelating agents, the washing efficiency abruptly decreased with the increase of pH as previously mentioned by several authors (Davies and Singh, 1995; Dikinya and Areda, 2010). The highest recovery was 83.5, 74.8, 85, 68.8, 88.7 and 72.5% for Cd, Co, Cu, Zn, Ni and Cr, respectively, which was achieved at very low pHe = 1.8.

With chelating agents, the washing efficiency was greatly improved under higher pH. In the presence of GLDA, target metals were efficiently removed at pHe = 3.3. At this pH, up to 83.9, 87.3, 81.2, 85.6, 89.3 and 90.2% of Cd, Co, Cu, Zn, Ni and Cr, respectively were successfully washed. While 70.7, 87.5, 72.9, 74.8, 86.2 and 77.6% were achieved at pHe = 4.2 for Cd, Co, Cu, Zn, Ni and Cr, respectively. The pH (3.3–4.5) corresponded to the middle and more controllable pH range. At pHe = 3.3, the removal efficiency could be classified as follows: Cr > Ni > Co > Zn > Cd > Cu. The high removal efficiency could be explained on one hand that all metals were not predominant in the residual form (F6) of the sequential fractionation, and on the other hand, it could be as a result of the stability of formed metal complexes with GLDA. In the previous study (Kołodynska, 2011a,b), the stability of metals towards GLDA at molar ratio GLDA: metal (1:1) was established as follows: Cu > Ni > Co $\approx$ Zn > Cd with respective stability constants as follows: 13.1 > 10.9 > 10 $\approx$ 10 > 9.1. This order can be compared to the one obtained in our results with slight difference, which could be justified by the different concentrations of metals within the sludge and the prevailing environmental conditions (competition effect between metals and other ions). Results from this study also show similarities to those reported by Wu et al. (2015). In their investigation, up to 89% Cd, 82% Ni and 84% Cu contents were recovered at the molar ratio of GLDA: metal = 3:1 at pH 4, whereas the removal efficiency of Zn remained low throughout the experiments. Similar to the presence of GLDA, metals were also better mobilized in the presence of CA at pHe ranging from 3 to 4.5. The extraction efficiencies at pHe = 3.3 for CA were 77.8, 74.8, 72.7, 65.8, 70.9 and 75% for Cd, Co, Cu, Zn, Ni and Cr, respectively. These removal efficiencies were lower when compared to those obtained using GLDA. Generally, target metals exhibited better extraction with GLDA than CA under different pH values.

The extraction of metals by organic ligands depends on the competition between the metal-binding functional groups from the sludge structure and the organic chelator (Wang et al., 2015). Development of high affinity of the metal towards a ligand will lead to the formation of a complex according to equation (1) bellow, where M^{n+} is a metallic ion and R-(COOH)$_m$ is an organic reagent (washing reagents). In the current study, compared with CA, GLDA exhibited stronger capability for metals removal from sludge. As a consequence, metals developed better affinity with GLDA than CA. This observation could be explained by its molecular structure with four carboxyl groups in comparison to CA with three carboxyl

Table 1
Metal sequential fraction of the original sludge.

Concentration (mg kg^{-1} dry sludge basis)						
Fractions	Cd	Co	Cu	Zn	Ni	Cr
F1	0.3	0.9	15.8	27.6	5.2	20.8
F2	0.7	0.2	56.7	43.4	15.7	101.8
F3	30.7	17.9	298.4	426.4	114.1	60.2
F4	10.1	14.1	67.8	340.7	32.3	82.8
F5	15.4	23.5	386.7	914.1	247.8	298.4
F6	1.4	5.2	104.1	221.9	34.2	31.6
$\sum$Fi	58.8	61.9	929.6	1974.2	449.4	595.7
TC	63.1	73.4	1103.2	2060.3	483.9	604.1
SEE (%)	**93.3**	**84.3**	**84.3**	**95.8**	**92.8**	**98.6**
RE (%)	**96.7**	**105.4**	**94.1**	**94.5**	**95.2**	**100.2**

Where TC is the total concentration of a given metal in sludge; $\sum$Fi is the sum of the different extracted fractions via sequential fractionation, SEE is sequential extraction efficiency in expressed as [($\sum$Fi)/TC] $\times$ 100; RE is the recovery obtained from reference sediment material.
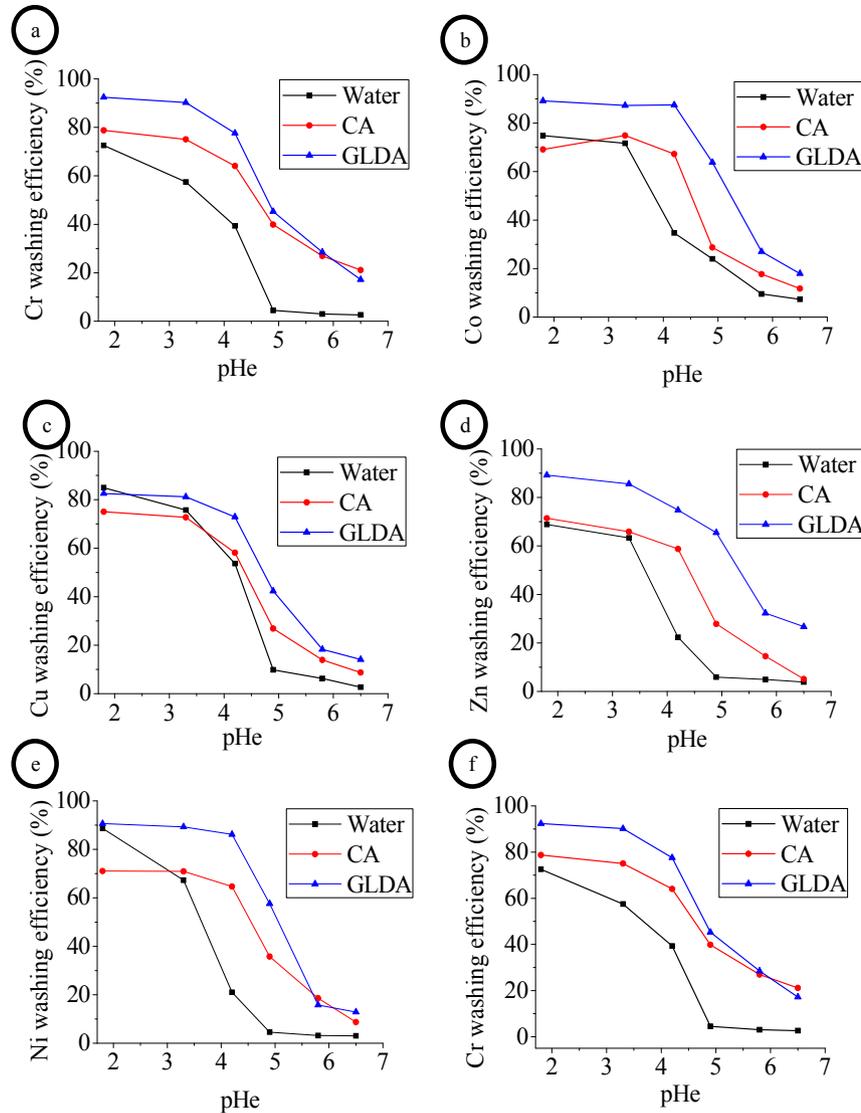
Fig. 1. Metals washing efficiency as function of pH (Conditions: 100 mM washing reagent and 12 h of washing).

groups (see Fig. S1 in SI). A molecule with more carboxyl groups has higher extracting capacity than a molecule with less carboxyl groups (Zaleckas et al., 2013).

$$\text{Sludge} - M^{n+} + R - (COOH)_m \rightarrow \text{Sludge} + R - Mn^+ (COOH)_m \tag{1}$$
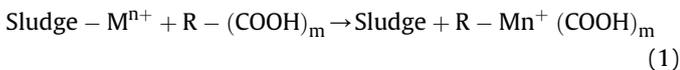
3.3. Effect of contact time on washing efficiency

The effect of contact time on extracting metals from sludge was investigated at the pHe = 3.3 using both CA and GLDA, and the results are shown in Fig. 2. From Fig. 2, it is deduced that when CA was used as the washing reagent, metals fully reached the state where no significant metal mobilization could be observed after 12 h of washing, and only 22.2, 25.2, 27.3, 34.2, 29.1 and 25.9% of Cd, Co, Cu, Zn, Ni and Cr, respectively remained in the sludge. When GLDA was used as the chelator, the equilibrium state was generally reached earlier. After 6 h of washing, only 22.9, 13.2, 18.8, 15.9, 13.2 and 9.2% of Cd, Co, Cu, Zn, Ni and Cr, respectively was left in the sludge. With GLDA, 6 h of washing was enough to achieve the best recovery efficiency for Co, Cu, Zn, Ni and Cr while another 6 h was needed to reach the equilibrium state for Cd. Longer washing

process after the equilibrium state did not increase the extraction efficiency. Therefore, for efficient mobilization of metals from sludge by CA and GLDA (100 mM), the sufficient contact time for the washing operation at pHe of 3.3 was about 6 h for GLDA and 12 h for CA. However, this time could vary depending on the forms of metals in the sludge as discussed below.

3.4. Effect of washing process on the distribution of metal forms in the sludge

In order to evaluate the change of metal distribution in the sludge and address the level of their stabilization, samples of both CA-washed and GLDA-washed sludge were subjected to sequential fractionation and results were compared to the original sludge (Fig. 3). Sequential fractionation of both CA-washed and GLDA-washed residues (pHe = 3.3) reveals considerable alterations in metal redistribution compared with the original sludge. In CA-washed residue (Fig. 3a), the most sensitive fractions (F1 and F2) were absent while carbonates bound proportion (F3) significantly decreased for all metals. The remaining metals were found bonded to the most stable fractions including Fe–Mn bound (F4), organic
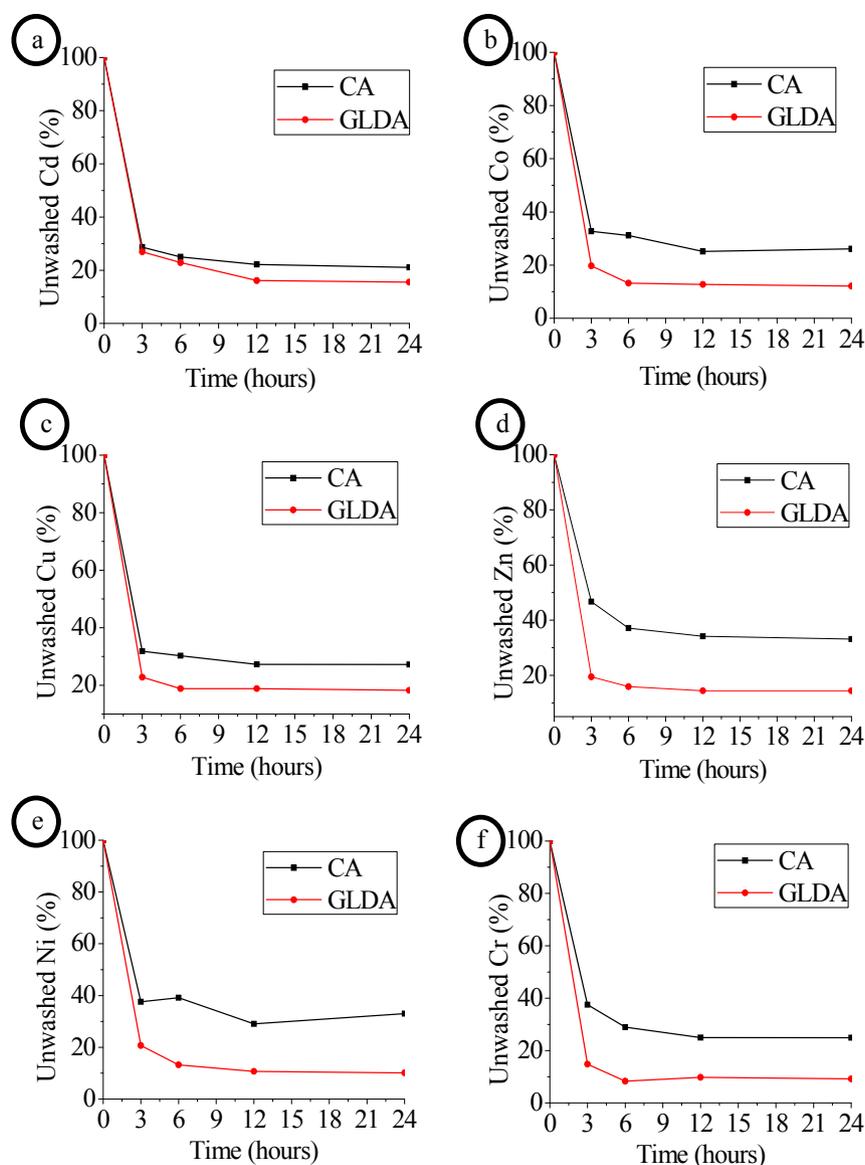
Fig. 2. Effect of contact time on washing efficiency (pHe = 3.3). (a) Cd, (b) Co, (c) Cu, (d) Zn, (e) Ni and (f) Cr.

matters-bound (F5) and residual (F6) fractions. In the case of GDLA-washed residue (Fig. 3b), metals were better stabilized compared with CA-washed residue. Accumulation of metals was not seen within F1, F2 and F3 and therefore, metals in all of the three fractions were completely washed, except that a relatively low concentration of Cd was detected in F3. This could be explained by the predominance of Cd within F3 in the raw sludge. Moreover, approximately all proportions of metals within F4 and F5 were washed for Co, Cu, Zn and Ni. Our results showed that nearly all metals remaining in GLDA-washed sludge were tightly bonded to the residual fraction (F6). Therefore, based on Fig. 3, it can be stated that all washed metals were from water soluble (F1), exchangeable (F2), carbonates bound (F3), Fe–Mn bound (F4) and organic matters bound (F5) fractions, and only silicate bound metals (residual metals), due to its refractory character, were irrecoverable.

3.5. Recovery of metals from leachate via hydroxide precipitation

Dissolved metals at pHe of 1.8 and under 24 h of washing conditions are 4.8, 5.2, 83.8, 147.2, 34.4 and 47.5 mg L^{-1} in CA-leachate and 5.2, 6.6, 91.7, 184.8, 44.1 and 56.1 mg L^{-1} in GLDA-leachate for Cd, Co, Cu, Zn, Ni and Cr, respectively. Results of hydroxide precipitation afterward are shown in Fig. S2 in SI. At pH 5.5, up to 79% and 73.5% of Cr in CA-leachate and GLDA-leachate, respectively were precipitated (Fig. S2a). However, the highest recovery for Cr was achieved at pH 7.5 (86.7%) for CA-leachate and 8.5 (82.4%) for GLDA-leachate. Co and Cu were better recovered at pH 8.5 with the efficiencies as follows: Co [CA-leachate (98.4%), GLDA-leachate (87.2%)] and Cu [CA-leachate (94.1%), GLDA-leachate (88.1%)]. Cd, Zn and Ni were best precipitated at pH 9.5 with the efficiencies as follows: [CA-leachate (94.7%), GLDA-leachate (87.5%)], [CA-leachate (96.3%), GLDA-leachate (85.6%)], and [CA-leachate (92.1%), GLDA-leachate (88.4%)], for Cd, Zn and Ni, respectively. Metal precipitation efficiency decreased beyond certain pH threshold, this implied the amphoteric characters of metals after reaching saturation pH. Otherwise, it can be noticed that metal recovery (precipitation) in CA-leachate seemed to be better than that in GLDA-leachate. This could be due to the high stability of metals complexes in the presence of GLDA as previously mentioned, and/or the higher concentration of metals in
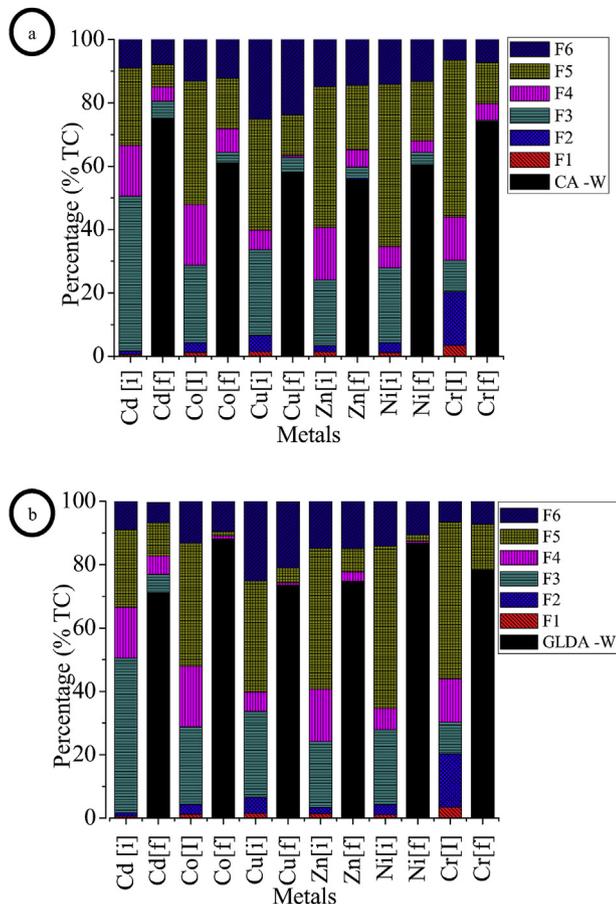
Fig. 3. Heavy metals fractionation in washed sludge (conditions: 24 h of washing, pHe = 3.3). (a) CA and (b) GLDA. [i] = initial, [f] = final, CA-W and GLDA-W respectively represent CA and GLDA washed portion of a given metal. TC is the total concentration of a given metal in the initial sludge.

GLDA-leachate as a result of better washing process of sludge.

3.6. Effect of pH and washing reagents on phosphorus (P) release

TP contents in the leachate from washing with water, CA and GLDA at different pH conditions are shown in Fig. 4. Results clearly show that P mobilization is pH dependent (Xu et al., 2015), and acidic conditions of the experiment favored P release. P mobilization decreased as pH increased towards the neutral pH. The highest release was achieved at the lowest pH = 1.8 with efficiencies of 54.8, 55.6 and 16.6% in the blank (water), CA and GLDA, respectively (Fig. 4). Besides, washing reagents also exercised remarkable
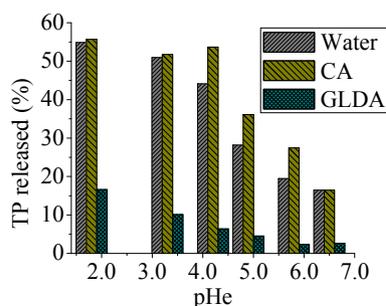
Fig. 4. Phosphorus released in washing leachate.

influence on P mobilization. While CA slightly increased P mobilization, GLDA totally exhibited an opposite effect. Unlike CA, GLDA tended to lower P solubilization and retained P within the sludge. This could be explained by the different nature of both compounds, for example, CA is an acidic compound while GLDA is alkaline. However, more research is needed to fully address the mechanism underlining this phenomenon. Since P is an important macronutrient for plants, less P release in the GLDA-washed sludge should be beneficial for the further application of the sludge in agricultural soils.

4. Conclusions

Comparative study of GLDA and CA for heavy metal extraction from sewage sludge was investigated. Results showed that compared with CA, GLDA possessed stronger and better capacity of removing metals from sludge. Both CA and GLDA considerably reduced the mobility of metals and greatly improved their stability in the washed sludge. However, unlike CA, GLDA tended to retain P within the sludge during the washing process, therefore making GLDA-washed sludge a better fertilizer for future agricultural use.

Acknowledgments

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvman.2015.10.035.

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