

Novel EDTA and process water recycling method after soil washing of multi-metal contaminated soil

Maja Pocięcha, Domen Lestān*

Agronomy Department, Biotechnical Faculty, University of Ljubljana, Jamnikarjeva 101, 1000 Ljubljana, Slovenia

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ABSTRACT

The development of EDTA-based soil washing technologies is hampered by the lack of treatment methods of the spent solution, particularly when multi-metal contaminated soils have to be remediated. Extraction of Pb (5329 mg kg⁻¹), Zn (3401 mg kg⁻¹), Cd (35 mg kg⁻¹) and As (279 mg kg⁻¹) contaminated soil with 60 mmol EDTA kg⁻¹ of soil removed 72%, 27%, 71%, and 80% of contaminants, respectively. We demonstrate here, on a laboratory scale experiment, the feasibility of using acid precipitation with HCl and H₂SO₄, coupled to initial alkaline Fe removal, to recover up to 88% of EDTA from a spent soil washing solution containing 11,578 mg L⁻¹ of EDTA and 1109, 267, 7.1 and 64 mg L⁻¹ of Pb, Zn, Cd and As, respectively. An electrochemical advanced oxidation process with a graphite anode was subsequently used to degrade 99.9% of the remaining EDTA in the spent washing solution and remove 99.7% Pb, 100% Zn, 96.6% Cd and 100% of As as (electro)precipitate. The cleansed process water obtained after electrochemical treatment was then used to prepare recycled washing solution by re-dissolving the recovered/recycled part of the EDTA. Washing solutions prepared from recycled EDTA had the same potential to extract Pb, Zn, Cd and As from soil as washing solution prepared from fresh EDTA of the same molarity. The novel recycling method is simple and robust and enables reuse of both EDTA and process water in a closed process loop.

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1. Introduction

The contamination of soils has become a major environmental concern. Remediation technologies can reduce the contamination and preserve soil as a non-renewable natural resource. Soil washing with chelants such as ethylenediamine tetraacetate (EDTA) permanently remove a variety of potentially toxic metals, metalloids and radionuclides (PTMs) from contaminated soils [1]. However, further development and commercialisation of EDTA-based soil washing is hampered by the large quantities of spent washing solution generated during the remediation process, which needs effective treatment before discharge into the environment or, better, recycling for reuse in the process. Furthermore, EDTA is toxic and poorly bio-degradable and thus poses a health and environmental hazard. Practical means of EDTA and process water recycling are still lacking, despite several treatment proposals. Demonstrated on a laboratory level, but not available commercially, current EDTA recycling involves substitution of toxic metals

in the EDTA complex and precipitation of the released metals in a pH gradient [2–5] or electric field [6–8].

It is especially difficult to treat spent washing solution containing several PTMs. Juang and Wang [7], for example, tested a membrane-separated, two-chamber electrochemical treatment for a binary, Pb and Cu, complexed EDTA solution and found lower metal recovery and current efficiency than for single metal contamination, regardless of the metal concentration ratios. We proposed electrochemical treatment of spent washing solution in a conventional single chamber electrolytic cell (without a membrane) under alkaline conditions (pH 10) using a sacrificial Al anode. We demonstrated the method feasibility for the treatment of spent washing solution containing a single toxic metal, Pb [8] and Cu [9] and their EDTA complexes. However, when we tried to optimise the method to treat multi-metal spent washing solution, up to 98% of Pb but only up to 73% of Zn and 66% of Cd were removed, while 88% of the recycled EDTA was preserved in the treated washing solution [10]. The problem is significant, since most sites are contaminated with several PTMs.

An approach that can be used efficiently to treat spent soil washing solutions containing several PTMs is first to degrade the EDTA and then to remove the released PTM ions by conventional precipitation and absorption methods. For example, in a combination of ozone and UV, an advanced oxidation process (AOP) generates hydroxyl radicals ($\cdot\text{OH}$) and has been used for effective oxidative

* Corresponding author at: Centre for Soil and Environmental Science, Biotechnical Faculty, University of Ljubljana, Jamnikarjeva 101, 1000 Ljubljana, Slovenia. Tel.: +386 01 423 1161; fax: +386 01 423 1088.

E-mail address: domen.lestan@bf.uni-lj.si (D. Lestān).

decomposition of EDTA in Pb and Zn complexes [11]. Hydroxyl radicals are very powerful, non-selective oxidants, second only to fluorine. The electrochemical AOP (EAOP) is more robust, technically simpler and cheaper. In the EAOP, hydroxyl radicals are produced at the anode according to Eq. (1), directly from the electrolysed water at a high current efficiency:



Wastewaters containing EDTA have been successfully treated with the EAOP [12,13]. The problems of this approach are the waste of EDTA, as a valuable chemical, and waste of energy for EDTA degradation.

The general purpose of our study is the need to reduce the overall cost of remediation treatment by soil washing, since the main item of cost of the treatment is the purchase of EDTA. In the current work, we examined the feasibility of a novel method for treating spent solution obtained after washing of Pb, Zn, Cd and As contaminated soil. The method combines the two general approaches outlined above: first, most of the EDTA is recovered and recycled from the spent washing solution using substitution and precipitation reactions in a pH gradient and the process water is then cleansed and recycled using the EAOP for oxidative degradation of the remaining EDTA and for PTMs separation. The efficiency of PTMs soil extraction using solutions with recycled EDTA was compared to the extraction efficiency of freshly prepared EDTA solution.

2. Materials and methods

2.1. Soil

Soil was collected from a vegetable garden in the Meza Valley, Slovenia. The valley has been exposed to more than three hundred years of active lead mining and smelting. The soil sample contained $5329 \pm 685 \text{ mg kg}^{-1}$ Pb, $3401 \pm 193 \text{ mg kg}^{-1}$ Zn, $35 \pm 6 \text{ mg kg}^{-1}$ Cd, $279 \pm 91 \text{ mg kg}^{-1}$ As and $42,628 \text{ mg kg}^{-1}$ Fe. For standard pedological analysis, the pH in the soils was measured in a 1/2.5 (w/v) ratio of soil and 0.01 M CaCl_2 water solution suspension. Soil samples were analyzed for organic matter by modified Walkley–Black titrations [14], cation exchange capacity (CEC) by the ammonium acetate method [15] and soil texture by the pipette method [16]. The following values were obtained: pH 6.57, organic matter 14.2%, CEC $20.7 \text{ mg } 100 \text{ g}^{-1}$ of soil, sand 51.0%, silt 42.5%, clay 6.5%. The soil texture was sandy loam.

2.2. Soil washing

The soil washing solution was obtained after extraction of 75 kg of soil with 75 L of $\text{Na}_2\text{-EDTA}$ solution (60 mM EDTA per kg of soil) in a concrete mixer for 2 h. The operating liquid:solid ratio did not significantly effect the efficiency of soil extraction (data not shown). After extraction, the soil suspension was first filtrated through a 2 mm sieve and the soil solid phase was then separated from the spent soil washing solution in a chamber filter press (filter cloth thickness 0.6 g cm^{-2} , air permeability $22 \text{ dm}^3 \text{ dm}^{-2} \text{ min}^{-1}$, Ecotip Ltd., Slovenia).

2.3. EDTA recovery

The spent soil washing solution was alkalized using 5 M NaOH up to pH 11.5 to precipitate Fe. The pH of the solution was then adjusted to values between 1 and 2 using 37% HCl or 96% H_2SO_4 to precipitate the EDTA. After shaking the alkalized and acidified solution for 2 h, Fe and EDTA precipitated and were removed from the solution by centrifugation at $3760 \times g$ for 10 min. The EDTA was dried at 60°C to a constant weight and stored as recycled EDTA

for further experiments. The residual solution was further electrochemically treated. The experiment was performed in triplicate.

2.4. Electrochemical treatment

Flow-through electrolytic cells (constructed from polyacrylamide) equipped with a graphite anode and stainless cathode (electrode surface 68 cm^2 , distance between electrodes 13 mm) were used. Washing solution (500 mL) was circulated from a magnetically stirred jar through the electrolytic cells using a peristaltic pump (flow rate 14 mL s^{-1}). A DC power supply (Elektronik Invent, Ljubljana, Slovenia) provided a constant electrical current density of 88 mA cm^{-2} . The initial pH of the solution was adjusted to pH 7 by drop-wise addition of 5 M NaOH. The contact time of the solution in the electrolytic cell was calculated as the ratio of cell volume and the volume of the washing solution, multiplied by the operation time (initially 19 min of operation time equalled 4 min of contact time). Ten mL samples were periodically collected and stored in the cold for PTM and EDTA analysis. The cathode was etched at the end with 30 mL of 65% HNO_3 to dissolve the electro-deposited PTMs. All the treatments were done in triplicate.

2.5. Efficiency of recycled EDTA

Washing solution with 30 mmol of recycled EDTA per kg of soil was prepared by dissolving the recovered EDTA (acid-precipitated as H_4EDTA) in deionised water or in the washing solution obtained after electrochemical treatment. A washing solution with an equimolar concentration of fresh $\text{Na}_2\text{-EDTA}$ in deionised water was also prepared. The pH of the washing solution with recycled EDTA was adjusted to pH 4.6. Solutions (20 mL) were then used for the extraction of contaminated soil (10 g) on a rotating shaker (1 h). After centrifugation at $2880 \times g$ for 10 min, the concentrations of Pb, Zn, Cd and As in the solutions were determined. Each extraction was done in triplicate.

2.6. EDTA determination

The concentration of EDTA was determined spectrophotometrically according to the procedure of [17]. The method involves the reaction of EDTA in washing solution with Fe^{3+} under acidic conditions to produce Fe-EDTA chelate, followed by the removal of excess Fe^{3+} by chelate extraction in the aqueous phase, using chloroform and N-benzoyl-N-phenylhydroxylamine and the formation of a chromophore with 4,7-diphenyl-1,10-phenanthroline-disulfonic acid. Using a spectrophotometer, absorbance was measured at 535 nm against a blank solution with the 4,7-diphenyl-1,10-phenanthroline-disulfonic acid replaced with an equal volume of distilled water. The lower limit of EDTA quantification was 20 mg L^{-1} .

2.7. Metal determination

Air-dried soil samples (1 g) were ground in an agate mill, sieved through a $160 \mu\text{m}$ mesh and digested in a microwave oven (CEM, MDS-2000) in 12 mL of *aqua regia* (20 min at 175°C) The digestions were conducted in triplicate. After cooling, digested samples were filtered through Whatman no. 4 filter paper and diluted with deionised water up to 100 mL. The pseudo-total concentrations of Pb, Zn, Cd and As were determined by flame (acetylene/air) AAS with a deuterium background correction (Varian, AA240FS). The metal concentration in the solutions was determined by AAS directly. A standard reference material used in inter-laboratory comparisons (Wepal 2004.3/4, Wageningen University, Wageningen, Netherlands) was used in the digestion and analysis as part of the QA/QC protocol. The limit of quantification for Pb, Zn, Cd, As and

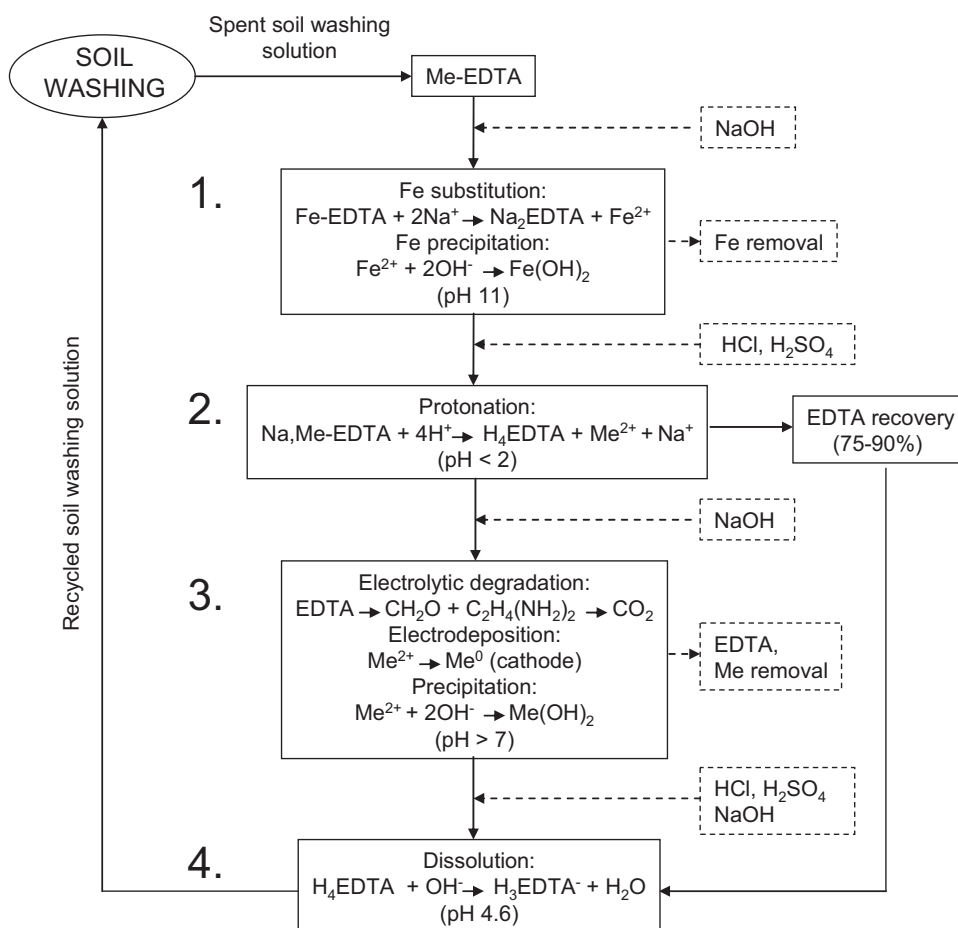


Fig. 1. EDTA and process water recycling scheme.

Fe were 0.1, 0.01, 0.02, 0.09 and 0.06 mg L⁻¹, respectively. Reagent blank and analytical duplicates were also used where appropriate, in order to ensure the accuracy and precision of the analysis.

2.8. Statistics

The Duncan multiple range test was used to determine the statistical significance ($p < 0.05$) between different treatments, using the computer program Statgraphics 4.0 for Windows.

3. Results and discussion

3.1. Extraction of Pb, Zn, Cd and As from soil using EDTA

In the initial soil extraction, we used 60 mmol kg⁻¹ EDTA to prepare spent soil washing solution for the further EDTA and process water recycling study. With this extraction, we removed 72%, 27%, 71%, 80% and 6.6% of Pb, Zn, Cd, As and Fe, respectively, from the contaminated soil. The concentration of EDTA, Pb, Zn, Cd, As and Fe in the waste soil washing solution was 11,578, 1109, 267, 7.1, 64 and 805 mg L⁻¹, respectively; the solution pH was 7.3.

As expected, Zn was the least extractable PTM. This had also been observed in our previous studies for soils from the same contaminated site. The low extractability was explained by the specific Zn association primarily with non-labile soil fractions [13]. The efficiency of As extraction with EDTA was surprisingly high, since EDTA does not form stable complexes with anionic metals and metalloids such as As [18]. However, EDTA can solubilise a significant amount of Fe from soil (see Fig. 2), probably from poorly crystalline Fe

oxyhydroxides. Oxyanions of As(V) (H₃AsO₄, arsenate) and As(III) (H₃AsO₃, arsenite) are the most abundant As species in soil, preferentially bound to Mn, Al and especially Fe oxides [19]. Mobilisation and removal of As from the soil could therefore be a consequence of partial solubilisation of Fe oxides.

3.2. EDTA and process water recycling

As shown in Fig. 1, the first step of the EDTA and process solution recycling process was the removal of Fe from the spent washing solution. Fe forms strong complexes with EDTA; the stability constant ($\log K_s$) of EDTA complex formation with Fe²⁺ is 14.3 (at 25 °C and ionic strength (μ) = 0.1) and with Fe³⁺ even 25.1 [20]. For comparison, the $\log K_s$ of other cationic PTM complexes with EDTA are 18.8, 16.5 and 16.4 for Pb, Zn and Cd, respectively [20]. Consequently, the initial soil extraction removed 6.6% of the total Fe soil content and the spent washing solution contained 805 mg L⁻¹ Fe (Fig. 2). Alkalinization of the spent soil washing solution to pH 11 and above removed Fe from the solution efficiently and completely. This was possible since, in alkaline solutions, Fe hydrolysis and Fe(OH)₃ precipitation make the formation of Fe-EDTA complexes less favourable. Under these conditions, Na or other alkali ion substituted Fe from the EDTA complex and the released Fe then precipitated as insoluble hydroxide [5].

The second step of the recycling process (Fig. 1) was acidification of the spent soil washing solution using HCl or H₂SO₄. EDTA is known to be poorly soluble in acidic water solution, from where it precipitates in its acidic protonised (H₄EDTA) form [21]. The efficiency of EDTA precipitation statistically significantly increased

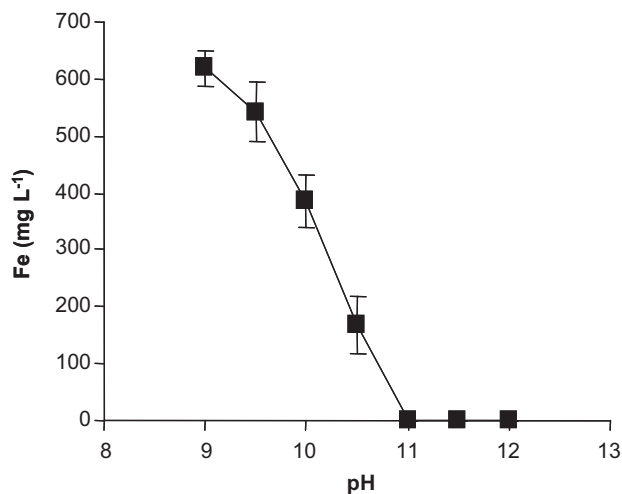


Fig. 2. Concentration of residual Fe in the spent soil washing solutions after removal of Fe by alkaline precipitation at different pH. Error bars represent standard deviation from the mean value ($n=3$).

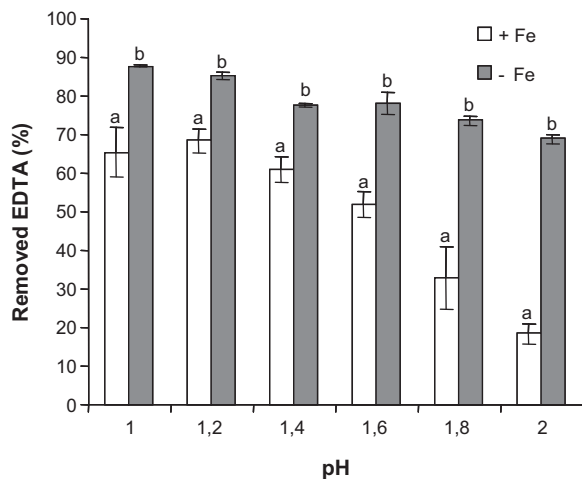


Fig. 3. Efficiency of reclamation of EDTA from spent soil washing solution at different pH values after acidification with HCl. Solutions with and without Fe were used in the experiment. Error bars represent standard deviation from the mean value ($n=3$). Letters (a and b) denote statistically different EDTA removal from solution according to the Duncan test ($p < 0.05$).

with a lowering of the pH of the spent washing solution (Fig. 3). The efficiency of EDTA precipitation and reclamation was also much higher (up to 88%) when Fe was removed from the spent washing solution prior to acidification (Fig. 3). For comparison, only up to 68% of initial EDTA was recovered from solutions containing Fe. The presence of Fe evidently interferes with EDTA protonation and precipitation reactions. A possible reason for this phenomenon is the stability of Fe–EDTA complexes, which is relatively high in acidic conditions between pH 1 and 2 and higher than the stability of EDTA complexes with other metals [22]. The interfering effect of Fe was even more pronounced when H_2SO_4 was used for acidification of the spent washing solution. Here, only up to 32% of EDTA precipitated in solution with Fe, compared to up to 78% of reclaimed EDTA from a solution without Fe (Fig. 4B).

Is initial Fe removal from spent soil washing solution always mandatory? Fe, namely, is not always reported to be present in these solutions in concentrations significant enough to expect interference with EDTA precipitation. While the $\log K_s$ of Fe–EDTA is high and the soil Fe concentration is usually higher than that of contaminating PTMs, the competitiveness of Fe species for

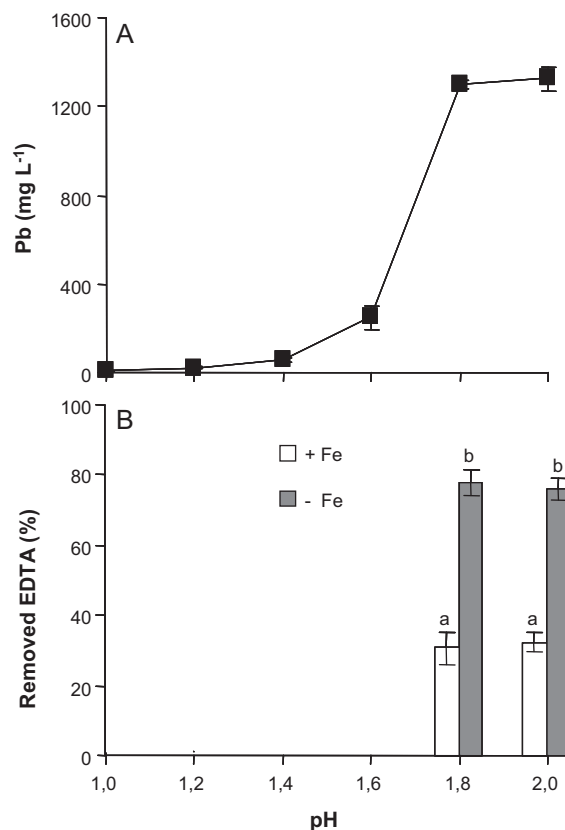


Fig. 4. Concentration of Pb in the spent soil washing solution (A) and the efficiency of reclamation of EDTA from solution at different pH values after acidification with H_2SO_4 (B). Solutions with and without Fe were used in the experiment. Error bars represent standard deviation from the mean value ($n=3$). Letters (a and b) denote statistically different EDTA removal from solution according to the Duncan test ($p < 0.05$).

complexation with EDTA might be hindered by the low aqueous solubility of Fe bearing soil minerals under usual soil conditions [23]. Borggaard [24] thus reported that EDTA extraction of Fe present as amorphous soil oxyhydroxides (i.e., goethite) was very slow and Elliot and Shastri [25] reported that EDTA did not extract Fe due to the coordination of several surface Ca ions by hexadentate EDTA, resulting in passivation of the Fe oxide surface. Information in the literature on Fe extractability is not univocal. Sun et al. [26], for example, reported that considerable mobilisation of Fe occurred during leaching of PTMs contaminated soil with EDTA. Furthermore, for contaminated sites in the Meza Valley we have reported that EDTA concentrations high enough to remove a significant part of PTMs also cause considerable Fe solubilisation [27]. Di Palma [28] reported that indigenous Fe was competitive with Cu in forming EDTA complexes and was removed from the soil by soil washing only when it was present in the organic and oxides–hydroxides fractions.

When H_2SO_4 was used for acidic EDTA precipitation, we observed parallel precipitation of Pb at $pH < 1.8$ (Fig. 4A). The stability of Pb–EDTA complexes decreases significantly in acidic pH [22]. Consequently, Pb was released from the EDTA and precipitated as insoluble $PbSO_4$ (other present contaminating PTMs do not form insoluble sulphates). Since co-precipitation of Pb and EDTA would contaminate the recycled chelant, only $pH \geq 1.8$ was used in further experiments. Applying H_2SO_4 for acidic EDTA precipitation at $pH \geq 1.8$ was otherwise significantly more efficient than using HCl in the same pH range (Duncan test, $p < 0.05$, Figs. 3 and 4B).

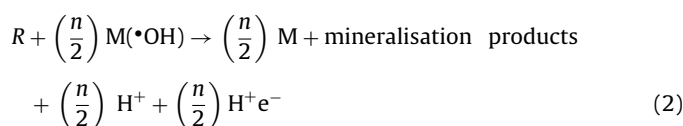
The third step (Fig. 1) was cleansing and reclamation of the spent washing solution as process water to prepare recycled

Table 1

Balance of Pb, Zn, Cd and As removal from soil washing solutions with EDTA precipitated by HCl and H₂SO₄ after EAOP treatment. The share (concentration) of PTMs electro-deposited on the cathode and in the solution were measured, the share of precipitated PTMs was calculated as the difference. Means ($n=3$) and standard deviation of results are presented.

	In solution (%)	Electrodeposited (%)	Precipitated (%)
Pb			
H ₂ SO ₄	2.5 ± 0.2	17 ± 3	80 ± 3
HCl	0.3 ± 0.3	15 ± 2	85 ± 2
Zn			
H ₂ SO ₄	13 ± 2	25 ± 4	61 ± 4
HCl	0	22 ± 1	78 ± 1
Cd			
H ₂ SO ₄	22 ± 2	19 ± 2	59 ± 2
HCl	3.4 ± 0.3	19 ± 0	77 ± 1
As			
H ₂ SO ₄	0	16 ± 5	84 ± 5
HCl	0	19 ± 3	81 ± 3

washing solution. After most of the EDTA was recovered (up to 88%, Fig. 3), the remaining EDTA, along with all the PTMs extracted from the soil, remained in the spent washing solution. To remove these residual contaminants, we applied electrochemical treatment in an electrolytic cell using the EAOP. In the EAOP, the electrode material is the most important parameter, since molecular oxygen is mainly produced on the anode during water electrolysis if the oxygen overvoltage on the anode is not sufficiently high. Materials such as graphite, Pt and various noble metal oxides on a Ti core have sufficient oxygen overvoltage to produce reactive hydroxyl radicals [29]. A boron-doped diamond anode (BDDA) has an extreme overvoltage of >3 V and has been proven to be particularly effective for oxidative degradation of EDTA [12,13]. BDDA, however, is still fairly expensive. In this study, we chose to use graphite, which is inexpensive, non-toxic and does not release metal ions back into the solution (to form undesirable EDTA complexes later on in the process). According to the generally accepted mechanism of EAOP, water is first discharged at the anode active sites (M), producing adsorbed hydroxyl radicals M(*OH), which are involved in the mineralisation of organic pollutants (R), in our case EDTA complexes with PTMs, in aqueous solution:



where (Eq. (2)) n is the number of electrons involved in the oxidation reaction of organics [30].

Selected process parameters in the electrolytic cell during treatment of soil washing solutions were measured. The final pH was 7.82 ± 0.02 and 7.42 ± 0.04 , respectively, for HCl and H₂SO₄ acidified solution. The range of voltage between the electrodes in the treatment with HCl acidified solution was from 7.3 to 9.0 V and in the treatment with H₂SO₄ solution from 8.0 to 11.3 V. Graphite consumption from the anode was $0.82 \pm 0.11 \text{ g L}^{-1} \text{ h}^{-1}$ (it was measured for the treatment with H₂SO₄ acidified solution only). As shown in Fig. 5, electrochemical removal of Zn and Cd, as well as EDTA, was slightly more effective from washing solution acidified with HCl. The final concentration of PTMs and EDTA in this washing solution was 2.7 ± 2.6 , 0.24 ± 0.02 and $6.1 \pm 1.2 \text{ mg L}^{-1}$ of Pb, Cd and EDTA, respectively. Concentrations of Zn and As were below the limit of quantification. Less than 20% of PTMs (except Zn) in solutions (both HCl and H₂SO₄ acidified) was removed by electrodeposition on the stainless steel cathode. The rest was precipitated from slightly alkaline media, probably as insoluble oxides and hydroxides (Table 1).

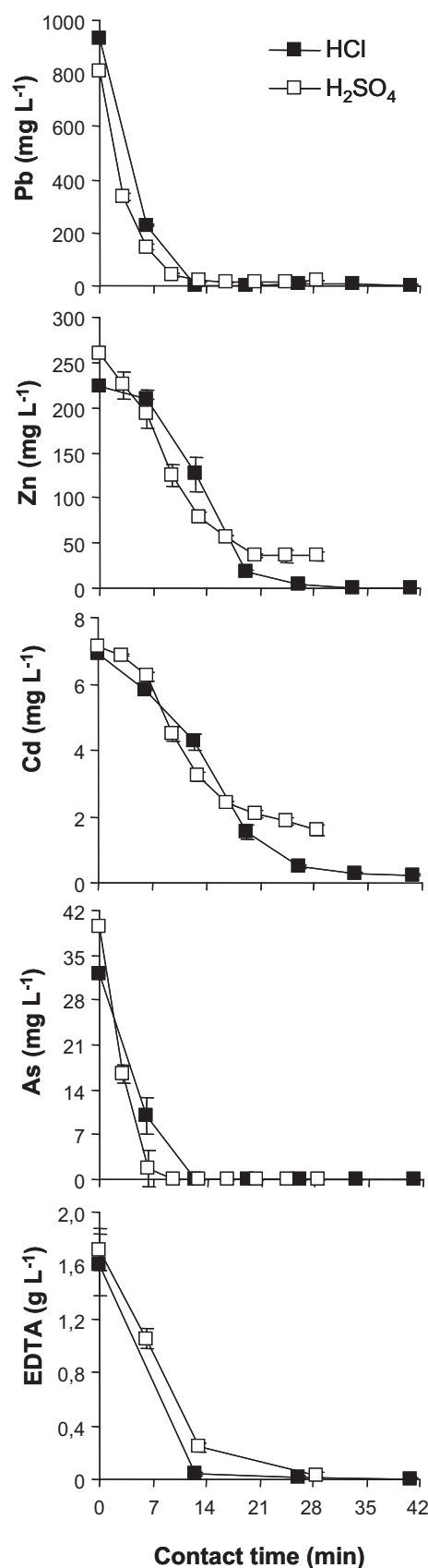


Fig. 5. Concentration of Pb, Zn, Cd, As and EDTA in the spent soil washing solution during EAOP. Solutions after acidic EDTA precipitation with HCl and H₂SO₄ were treated. Error bars represent standard deviation from the mean value ($n=3$).

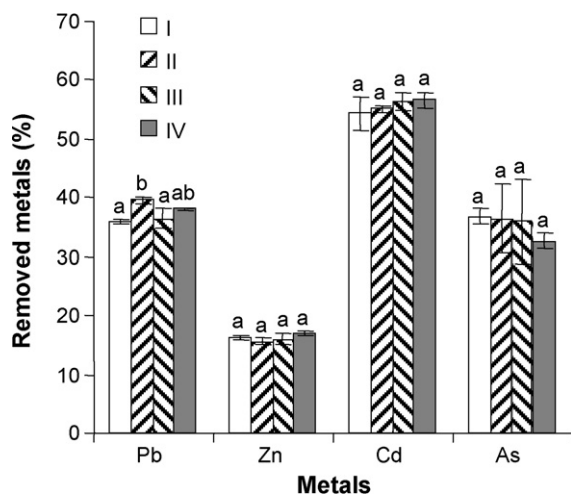


Fig. 6. Efficiency of Pb, Zn, Cd and As removal from soil using (I) fresh EDTA in deionised water, (II) recycled EDTA (H_2SO_4 precipitated) in deionised water, (III) recycled EDTA (HCl precipitated) in deionised water and (IV) recycled EDTA (HCl precipitated) in reused cleansed process water. The EDTA concentration was 30 mmol kg^{-1} of soil in all solutions. Error bars represent standard deviation from the mean value ($n=3$). Letters (a and b) denote statistically different PTM removal from solution according to the Duncan test ($p < 0.05$).

In relation to the EDTA degradation pathway in the electrolytic cell, Johnson et al. [31] reported that a Pt anode oxidized EDTA into CO_2 , formaldehyde (CH_2O) and ethylenediamine ($\text{C}_2\text{H}_4(\text{NH}_2)_2$). Yamaguchi et al. [32] reported that EDTA was electro-oxidized through sequential removal of the acetate groups, until unidentified small size hydrocarbon products were formed.

In the final, fourth step (Fig. 1), we prepared recycled soil washing solution by dissolving the EDTA reclaimed and recycled by acidic precipitation, in the cleansed process water obtained after the EAOP. The recycled EDTA was present in protonated H_4EDTA form and was presumably dissolved through increasing stages of EDTA deprotonation and complex formation with cations present in the solution (e.g., Na^+). For the purpose of further testing the PTM soil extraction potential of the recycled EDTA (reclaimed using HCl or H_2SO_4) H_4EDTA was also dissolved in deionised water. The pH of prepared washing solutions with recycled EDTA was adjusted to pH 4.6, which was the pH of the solution with fresh EDTA.

3.3. Extraction of PTMs from soil using recycled and fresh EDTA

Except for slight differences in Pb removal, the results indicate no statistically significant differences between the efficiency of recycled EDTA solutions (precipitated using HCl and H_2SO_4) and freshly prepared EDTA solution, in extracting PTMs from soil (Fig. 6). Whether the recycled EDTA was dissolved in deionised water or in reused cleansed process water also did not affect Pb, Zn, Cd and As extraction (Duncan test, $p < 0.05$). Using 30 mmol kg^{-1} EDTA solutions removed up to 40%, 17%, 56% and 36% of Pb, Zn, Cd and As, respectively, from contaminated soil. Using higher 60 mmol kg^{-1} EDTA removed much more PTMs (Section 3.1) but further increasing the EDTA concentration soon afterwards reached a plateau in terms of the percentage of removed PTMs (data not shown). With this in mind, we chose to use a somewhat lower chelant concentration, in order better to highlight possible differences in the potential of recycled and fresh EDTA to extract PTMs.

In the further development of EDTA-based soil washing technologies after soil extraction the soil rinsing with clean solution must be considered to remove the toxic residual EDTA and EDTA mobilised PTMs from remediated soil. This will generate additional

volumes of used solution, which will need effective treatment before discharge or reuse.

4. Conclusions

The results of our laboratory scale study indicate the feasibility of the novel method of EDTA and process water recycling, as part of the washing technology for multi-metal contaminated soils. The following specific conclusions can be drawn:

- Alkaline precipitation and removal of Fe prior to acidic EDTA precipitation greatly enhances the efficiency of chelant reclamation.
- Using HCl for acidic EDTA precipitation at very low pH yields the highest percentages of chelant reclamation.
- PbSO_4 precipitation limits the use of H_2SO_4 for acidic EDTA precipitation to $\text{pH} > 1.8$. In this pH range, however, H_2SO_4 is a more efficient EDTA precipitant than HCl.
- The residual EDTA and contaminating metals can be efficiently removed from the spent soil washing solution in an electrolytic cell using a chemically inert graphite anode.
- The washing solution prepared from recycled process water and recycled EDTA is equally effective in the removal of Pb, Zn, Cd and As from contaminated soil as washing solution prepared from fresh water and fresh EDTA.
- Recycled and fresh EDTA washing solutions efficiently removed As (anionic metalloid) from soil, presumably by dissolution of arsenate and arsenite bearing Fe oxides.

The novel treatment method for spent soil washing solution is robust and characterized by simple equipment, a brief retention time and easy operation.

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