

# Investigation of different ethylenediamine-*N,N'*-disuccinic acid-enhanced washing configurations for remediation of a Cu-contaminated soil: process kinetics and efficiency comparison between single-stage and multi-stage configurations

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**Abstract** A comparison of Cu extraction yields for three different ethylenediamine-*N,N'*-disuccinic acid (EDDS)-enhanced washing configurations was performed on a Cu-contaminated soil. Batch experiments were used to simulate a single-stage continuous stirred tank reactor (CSTR) and a multi-stage (side feeding and counter-current) reactor. Single-stage CSTR conditions were simulated for various EDDS:(Cu + Cd + Pb + Co + Ni + Zn) molar ratio (EDDS:M ratio) (from 1 to 30) and liquid to soil (L/S) ratio (from 15 to 45). The highest Cu extraction yield ( $\approx 56\%$ ) was achieved with EDDS:M = 30. In contrast, a Cu extraction yield decrease was observed with increasing L/S ratio with highest extracted Cu achievement ( $\approx 48\%$ ) for L/S = 15. Side feeding configuration was tested in four experimental conditions through different fractionation mode of EDDS dose and treatment time at each washing step. Results from the four tests showed all enhanced Cu extraction (maximum values from  $\approx 43$  to  $\approx 51\%$ ) achieved at lower treatment time and lower EDDS:M

molar ratio compared to CSTR configuration with L/S = 25 and EDDS:M = 10. The counter-current washing was carried out through two washing flows achieving a process performance enhancement with 27% increase of extracted Cu compared to single-stage CSTR configuration. Higher Cu extraction percentage (36.8%) was observed in the first washing phase than in the second one (24.7%).

**Keywords** EDDS-enhanced soil washing · Cu extraction yield · CSTR configuration · Multi-step washing · Side feeding configuration · Counter-current configuration

## Introduction

Soil contamination by heavy metals (HMs) is a worldwide problem (Finzgar and Lestan 2007), and their excessive accumulation leads to health risks to animals and human beings besides having deleterious effects on soil fertility and ecosystem functions (Sun et al. 2001). This led scientific and engineering communities to look for suitable soil remediation technologies (Guo et al. 2010). Among several strategies, the soil washing approach showed promising efficacy especially if enhanced by the use of chelating agents (Ferraro et al. 2016a). Among these, several studies focused on the use of aminopolycarboxylic acids (APCs) that are characterized by many advantages such as high metal extraction efficiency, high metal complex stability and solubility, and low APC adsorption phenomena onto soil particles (Fischer et al. 1998). Furthermore, the involvement of biodegradable chelating agent such as ethylenediamine-*N,N'*-disuccinic (EDDS) in soil washing entailed the implementation of more environmental friendly and safe remediation process (Hauser et al. 2005; Tandy et al. 2006).

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Nonetheless, soil washing process can be an expensive remediation technique when high amount of contaminated soils needs to be treated, thus limiting process feasibility to the remediation of smaller contaminated areas. Also, the high cost of extracting agents as well as the required treatment of spent washing solution for its safe disposal is seen as an obstacle for full-scale applications (Lim et al. 2005). For this reason, several technical improvements are required in order to limit the process operating costs. For instance, involvement of physical separation techniques in pilot- and full-scale soil washing units are useful to concentrate metal contaminants in smaller soil volume (Dermont et al. 2008). In addition, different recovery techniques, such as chemical precipitation, washing solution evaporation and acidification, and electrochemical treatment involving two- or single-chamber cell, were investigated to reuse the spent extracting solution in further washing steps (Allen and Chen 1993; Ager and Marshall 2003; Di Palma et al. 2003; Zeng et al. 2005; Di Palma et al. 2005; Voglar and Lestan 2010; Pocięcha and Lestan 2010; Ferraro et al. 2015).

Besides these different techniques aiming at lowering soil washing procedure costs, proper soil washing configuration could represent another operating parameter to consider for process cost-effectiveness optimization. Previous comparison between single CSTR and multi-step soil washing already displayed process efficiency enhancement when several washing steps were involved (Steele and Pichtel 1998). However, comparison of contaminant extraction kinetics and optimal operational condition selection (i.e., treatment time, chelating agent dose, liquid to soil ratio) for different configurations are still absent in the available literature.

The aim of this study is to simulate at lab-scale different EDDS-enhanced washing reactor configurations on an agricultural Cu-contaminated soil and investigate the best conditions for efficient contaminant extraction. EDDS was selected as extracting agent for the well-asserted applicability in the HM-contaminated soil remediation (Race et al. 2016; Satyro et al. 2016) and its high biodegradability (Fabbricino et al. 2013). The investigated process configurations are the commonly applied single-stage CSTR configuration and multi-stage washing simulated in two different configurations: (i) side feeding (SF) plug flow and (ii) counter-current (CC) washing (Levenspiel 1972). Single-stage CSTR tests allowed to determine suitable process parameters (e.g., EDDS dose, liquid to soil ratio, treatment time) for the achievement of the highest Cu extraction efficiency. These parameters are implemented in SF and CC configuration tests keeping constant the total amount of used EDDS, the total reaction time, and the total amount of treated soil. The outcomes from the tests are used to assess the best combination of EDDS dose/treatment time fractionation and washing configuration in order to improve the soil washing process in terms

of Cu extraction efficiency. Data from multi-stage configurations are compared with single-stage CSTR in terms of process kinetics and extraction efficiency. The aim is to investigate the possibility to minimize the EDDS amount used and operational time involved. This could possibly result in economical/technical benefits for soil washing full-scale applications.

## Materials and methods

### Soil physical-chemical characteristics

The experimental activities were carried out with a Cu-contaminated soil previously devoted to agricultural use and collected in Castel San Giorgio in the south of Italy. Soil samples were collected up to a depth of 50 cm from soil layer surface. Soil pH and electrical conductivity (EC) were determined using a pH meter (Orion 420A<sup>+</sup>, Thermo) and an EC meter (XS Cond 6), respectively, in a mixture with a soil:distilled H<sub>2</sub>O ratio = 1:2 (w/v). ASTM methodologies were used to evaluate the soil particle size distribution (ASTM D 422-63 2007), initial field moisture, and organic matter (ASTM D 2974-00 2000). Soil samples were heated at 105 °C until constant weight for the initial field moisture content determination. Organic matter was obtained by loss-on-ignition test at 550 °C in a muffle furnace.

Ammonium acetate method was used to evaluate potential cation exchange capacity (CEC) (Espino-Mesa et al. 1993). This method was implemented on 4 g of soil sample through leaching 33 ml of 1 M NH<sub>4</sub>OAc solution (pH = 7) for 5 min. An ethanol solution (95% concentrated) was used for soil wash after NH<sub>4</sub>OAc solution leaching followed by three washes of 33 ml 1 M KCl solution. Concentration of adsorbed NH<sub>4</sub><sup>+</sup> was evaluated through titrimetric method and used to calculate potential CEC.

Soil acid digestion was conducted through microwave-assisted acid digestion (USEPA 1995) using a Milestone START D microwave oven for the evaluation of total HM content.

Characterization in terms of HM fractionation in soil was performed through a modified BCR (Community Bureau of Reference) three-step sequential extraction (Pueyo et al. 2008) to determine the Cu concentration in the non-detrital and detrital fraction of the soil. The non-detrital fraction is composed by carbonates (exchangeable and weak acid soluble fraction) and Fe-Mn oxides (reducible fraction) characterized by less strong bond with HMs than detrital fraction that is composed by organic matter and sulfide (oxidizable fraction) and mineral matrix constituents (residual fraction) (Komárek et al. 2008). The above-cited soil properties are reported in Tables 1, 2, and 3.

**Table 1** Physical properties of the soil

Initial field moisture <sup>a</sup> (%)	Volatile solids <sup>a</sup> (g kg <sup>-1</sup> )	Electrical conductivity <sup>b</sup> (μS cm <sup>-1</sup> )	Soil pH <sup>b</sup>	Cation exchange capacity <sup>c</sup> (meq/100 g of soil)
20 ± 1.29	75.64 ± 2.06	430.67 ± 9.29	7.81 ± 0.2	29.56 ± 0.44

<sup>a</sup> Standard test methods for moisture, ash, and organic matter of peat and other organic soils (ASTM D 2974-00 2000)

<sup>b</sup> Soil:ultra pure H<sub>2</sub>O ratio = 1:2

<sup>c</sup> Method for actual and potential cation exchange capacity (CEC) determination (Espino-Mesa et al. 1993)

### Lab-scale soil washing tests

Prior each soil washing test, the contaminated soil samples were sieved at 2 mm to remove gravels from the studied soil samples. All calculations were carried out referring to the soil dry weight. The initial soil moisture was removed by air-drying the investigated soil before each test. (S,S)-ethylenediamine-*N,N'*-disuccinic acid trisodium salt solution (35% in H<sub>2</sub>O) for washing solution preparation was provided by Sigma-Aldrich. Proper amount of EDDS was mixed with distilled (DI) water for each set of experiments according to the EDDS:M molar ratio established for the specific test. The term M is referring to the sum of Cu, Cd, Pb, Co, Ni, and Zn moles per kg of soil, while the EDDS:M molar ratio was calculated by evaluating the molar amount of EDDS per kg of soil against the molar amount of metals per kg of soil.

Soil washing tests were performed in 500-ml glass beaker reactors and total washing solution volume of 400 ml. Proper mix of soil and EDDS solution was obtained through an ISCO jar test equipment at 150 round per minutes (rpm). The reactors were closed by Parafilm® layer on the top in order to avoid the EDDS solution evaporation and alteration of the selected L/S ratio during the treatment time. EDDS washing solution pH was monitored all along the tests displaying values of 8.0 ± 0.2 that represented the natural pH of the investigated soil.

All the collected slurry samples were centrifuged using an IEC CENTRA GP8R centrifuge at 4600 rpm for 20 min to separate the treated soil from the liquid phase. The latter was then filtered through 0.45-μm fiberglass filters to remove residual soil particles and stored at 4 °C until analysis.

### Single-stage CSTR washing configuration

Single-stage CSTR configuration was simulated through single washing step tests by accurately mixing contaminated soil

**Table 2** Particle size distribution of the soil

Clay <sup>a</sup> (%)	Silt <sup>a</sup> (%)	Sand <sup>a</sup> (%)	Gravel <sup>a</sup> (%)
16.24 ± 0.97	44.03 ± 1.54	32.59 ± 1.06	7.14 ± 0.48

<sup>a</sup> Test method for particle size analysis of soils (ASTM D 422-63 2007)

and extracting solution with jar test equipment at 150 rpm. A long treatment time of 96 h was implemented in order to reach asymptotic condition regarding Cu extraction efficiency. About 10 ml samples were collected during the experiment using a 50-ml plastic syringe at 1, 3, 6, 18, 24, 48, and 96 h and stored at 4 °C until analysis. Different values of EDDS:M molar ratio and L/S ratio were investigated to optimize the operational parameters. Experiments with different molar ratio were performed at EDDS:M molar ratio of 1, 10, 20, 30, and L/S ratio fixed at 15 (v/w). EDDS:M ratio values were selected to have a wide range of investigation on process efficiency varying from stoichiometric value to high EDDS moles excess compared to the total HMs moles. EDDS:M ratio was calculated as EDDS moles per sum of initial Cu, Cd, Pb, Co, Ni, and Zn moles in soil. This latter was evaluated analyzing solution from soil acid digestion test, and suitable EDDS concentrations were considered accordingly in order to have precise EDDS moles to achieve the desired EDDS:M ratio values. EDDS:M ratio calculation was limited to the soil contaminants in order to have sufficiently available EDDS moles for Cu extraction and avoid competition phenomena between these contaminants for EDDS chelation. In particular, the present study focused on the Cu extraction as main contaminant in the investigated soil. Regarding the tests performed with different L/S ratio, the investigated values were 15, 25, 35, and 45 with EDDS:M ratio fixed at 10. The L/S ratio was varied by changing soil amount in the test and keeping washing solution volume constant and equal to 400 ml. As well as for EDDS:M ratio values, also the selection of L/S ratio values was made in order to investigate variability of process efficiency in a wide range of different soil amounts (from 26.7 to 8.9 g).

### Multi-stage washing configurations

The multi-stage washing configurations were simulated by performing multi-washing steps. EDDS:M ratio = 10 and L/S ratio = 25 were selected from the single-stage CSTR tests according to the significant extracted Cu percentage achieved under these conditions. EDDS molarity of 3.14 mM corresponded to a EDDS:M ratio = 10 with a total EDDS dose of 1.256 mmol in 400 ml of washing solution in the single-

**Table 3** Investigated element total concentration in the soil

Total Cu <sup>a</sup> (mg kg <sup>-1</sup> )	Total Cd <sup>a</sup> (mg kg <sup>-1</sup> )	Total Pb <sup>a</sup> (mg kg <sup>-1</sup> )	Total Co <sup>a</sup> (mg kg <sup>-1</sup> )	Total Ni <sup>a</sup> (mg kg <sup>-1</sup> )	Total Zn <sup>a</sup> (mg kg <sup>-1</sup> )	Total Fe <sup>a</sup> (mg kg <sup>-1</sup> )	Total Ca <sup>a</sup> (mg kg <sup>-1</sup> )	Total Mg <sup>a</sup> (mg kg <sup>-1</sup> )	Total Mn <sup>a</sup> (mg kg <sup>-1</sup> )
167.5 ± 7.19	0.743 ± 0.04	74.81 ± 9.85	7.73 ± 0.65	61.88 ± 2.72	125.89 ± 13.19	33,178.6 ± 40.47	56,138.1 ± 92.11	483.4 ± 22.57	459.2 ± 16.99

<sup>a</sup> Microwave-assisted digestion of sediments, sludges, soils, and oils (USEPA 1995)

stage test. Total treatment time of 24 h was selected, corresponding to the final value before the occurrence of the Cu extraction asymptotic condition observed in the single-stage CSTR experiment. SF and CC configurations were then performed fractionating the total treatment time and total EDDS dose in eight washing steps. The total washing solution volume of 400 ml involved in the single-stage CSTR test was also fractionated in the eight washing steps to have a proper configuration comparison also in volumetric terms.

SF configuration tests were carried out by involving 50 ml of washing solution in each washing step on a soil total amount of 2 g in order to have a L/S ratio of 25. Fresh EDDS solutions were involved in each step, and four different washing conditions were simulated: (A1) constant EDDS dose (0.158 mmol) and treatment time (3 h) for each washing step, (A2) increasing EDDS dose (0.049, 0.080, 0.111, 0.142, 0.172, 0.203, 0.234, and 0.265 mmol) and constant treatment time (3 h) for each washing step, (A3) decreasing EDDS dose (0.265, 0.234, 0.203, 0.172, 0.142, 0.111, 0.080, and 0.049 mmol) and constant treatment time (3 h) for each washing step, and (A4) constant EDDS dose (0.158 mmol) and increasing treatment time (0.5, 1, 1.5, 2, 2.5, 3.5, 5, and 8 h) for each washing step. A1, A2, and A3 washing conditions were selected in order to study all possible EDDS dose fractionation modalities. Table 4 summarizes the experimental setup of the above reported washing conditions.

CC configuration was studied by using 0.25 g of soil for each of eight washing steps, resulting in a total amount of treated soil of 2 g. This was useful to properly compare cumulative extracted Cu results with previous washing configurations by considering the same operational conditions. The eight soil fractions were defined as S1, S2, S3, S4, S5, S6, S7, and S8 and were sequentially washed by two fresh EDDS solutions. The first EDDS solution performed the eight washing steps from S1 to S8 (first washing phase), while the second EDDS solution performed the eight washing steps from S8 to S1 (second washing phase) in order to simulate counter-current washing condition. The EDDS dose was equally divided between the washing solutions (0.628 mmol) of 200 ml each. Constant treatment time equal to 1.5 h was selected for each washing step in order to have a total time of 24 h.

### Glassware preparation and analytical methods

All the glassware involved for solution preparation and soil washing experiments was stored in 2% HCl bath overnight and rinsed with DI water prior use. All samples collected from soil acid digestion and soil washing tests were analyzed through atomic adsorption spectrometry (AAS) using a Varian Model 55B SpectrAA (F-AAS) equipped with a flame (acetylene/air) and a deuterium lamp for background correction to determine initial concentration of Cu, Cd, Pb, Co, Ni, and Zn and extracted Cu during washing tests.

The reported data were averaged from triplicate experiment results, and corresponding standard deviations were shown as error bars.

## Results and discussion

### Single-stage CSTR washing configuration

HM and main cation initial concentrations were determined from extracting solution of soil acid digestion test. Results showed initial concentrations as reported in Table 3.

Figure 1a shows the cumulative extracted Cu percentage after each sampling time with various molar ratios. As observed in a previous study (Ferraro et al. 2016b), two main extraction kinetic paths can be distinguished as the reaction time increases. A Cu fast extraction occurred in the early hours of the treatment followed by an extraction rate declining up to occurrence of no significant extracted Cu percentage increase. Besides the treatment time that highly affects the effectiveness of the extraction process (Vandevivere et al. 2001), it is also reported that heavy metal speciation into the soil can affect the contaminant mobility and solubilization by chelating agents used (Elliott et al. 1989).

Generally, the sum of non-detrital fractions (e.g., exchangeable, carbonate, and reducible fractions) determines the metal amount that can be extracted by soil washing process enhanced by chelating agents (Peters 1999). Then, weakly bound metals undergo to an initial faster extraction rate that is followed by a slower release of the metals that are more strongly bound to the soil (Abumaizar and Smith 1999).

As observed in a previous work (Ferraro et al. 2015), sequential extraction results showed that only 2.2 and 15.8% of Cu were present in the exchangeable fraction and reducible fraction, respectively, in the studied soil, while 50.7% were present in the oxidizable fraction and 31.3% in the residual fraction. According to this, only the test with EDDS:M ratio equal to 1 showed an extracted Cu percentage in the early treatment hours corresponding to the weakly bound Cu fraction percentage.

In contrast, the increase of EDDS:M ratio displayed a higher extraction efficiency than expected from the beginning of the treatment. This could be mainly ascribable to metal mobility enhancement due to the higher concentration of chelating agents (Elliott et al. 1989; Abumaizar and Smith 1999; Sarkar et al. 2008). Another factor influencing the metal extraction efficiency can be the release of cations (e.g., Ca and Fe) coexisting with soil contaminants due to the soil matrix constituent dissolution (Steele and Pichtel 1998). As a consequence, competition phenomena for the chelant complex formation can occur between main cation competitors and HMs, reducing the amount of free ligand available to chelate the soil



**Table 4** SF configuration experimental setup

Washing conditions	EDDS dose (mmol/step)	Treatment time (h/step)	Washing steps
A1	Constant (0.158)	Constant (3)	8
A2	Increasing (0.049, 0.080, 0.111, 0.142, 0.172, 0.203, 0.234, 0.265)	Constant (3)	8
A3	Decreasing (0.265, 0.234, 0.203, 0.172, 0.142, 0.111, 0.080, 0.049)	Constant (3)	8
A4	Constant (0.158)	Increasing (0.5, 1, 1.5, 2, 2.5, 3.5, 5, 8)	8

contaminants (Kim et al. 2003; Koopmans et al. 2008; Subirés-Muñoz et al. 2011).

Competition phenomenon effects can be more noticeable at low chelant:HM molar ratio, especially for ratio lower than 1 (Begum et al. 2012). Therefore, high chelant mole excess is

required in order to increase HM extraction from soil (Tandy et al. 2004; Lestan et al. 2008).

Figure 1b reports the Cu extraction efficiency for L/S ratio values ranging from 15 to 45. In contrast to the results observed by varying the EDDS:M ratio, it was possible to notice that extracted Cu percentage was decreasing with increasing L/S ratio values. It is reported that L/S ratio increase can generally entail positive effect on the soil washing process efficiency (Zou et al. 2009). This result is ascribed to the increase of chelant:HM molar ratio with increasing L/S ratio when chelant molarity in solution is kept constant (Zou et al. 2009).

In contrast, further works also reported that slight effect on the extraction efficiency occurred while varying L/S ratio or mainly no effect was observed with constant value of chelant:HM molar ratio and variable L/S ratio (Van Benschoten et al. 1997; Tsang et al. 2012). In the present study, the increase of L/S ratio was achieved through soil amount decrease with constant values of EDDS:M ratio and liquid phase volume. As a consequence, decreasing moles of HMs and EDDS were obtained for L/S ranging from 15 to 45. This likely led to higher chelation competition occurrence at the lowest EDDS mole values as already observed from results with different EDDS:M ratio (Fig. 1a).

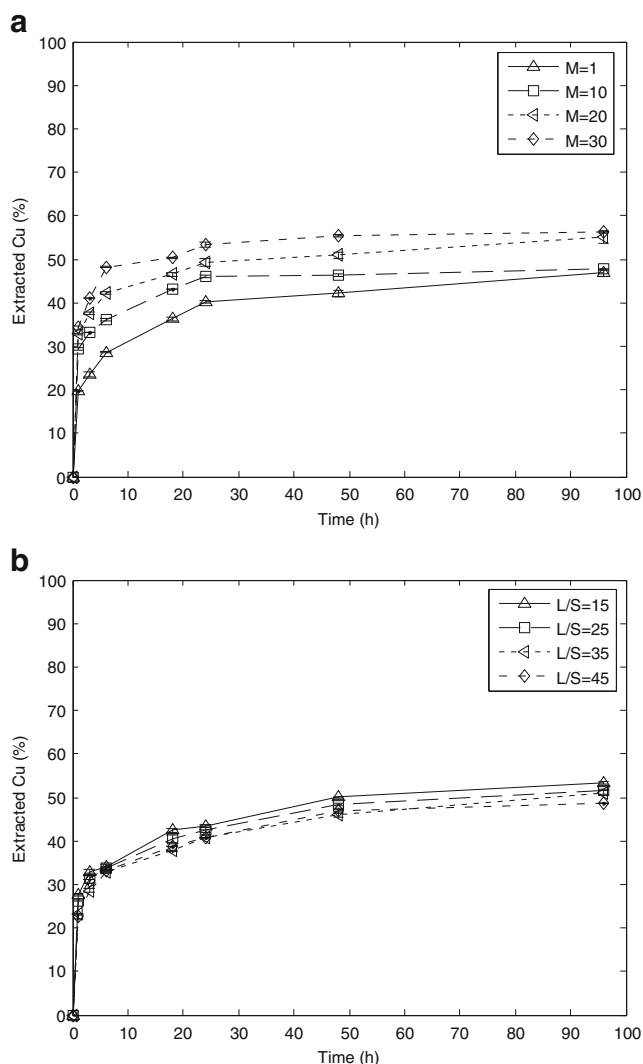
However, Fig. 1b displays similar Cu extraction percentages among all tests occurring after 24 h of treatment time. These latter results suggested that increasing L/S ratio values were mainly delaying the washing process, resulting in longer treatment time necessary to achieve suitable extraction efficiency.

**Side feeding washing configurations**

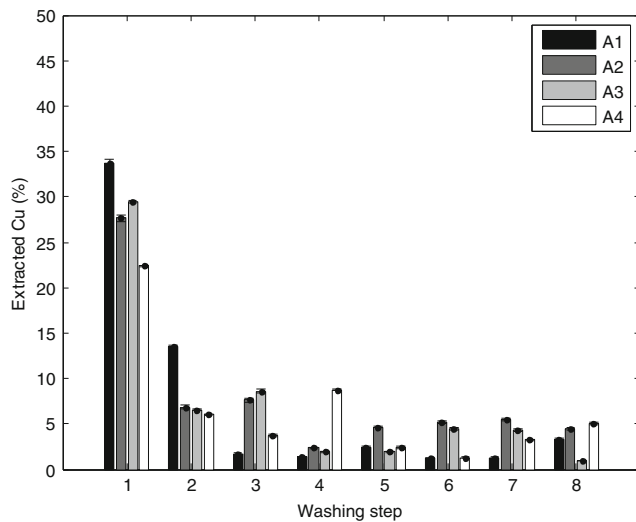
*Cu extraction yield*

Results obtained from the four experimental setups of the SF washing configuration are reported in Fig. 2.

In the various tests, one can observe some slight fluctuations in terms of extracted Cu percentage from washing steps 3 to 8. This can be ascribed to Cu mobilization from a washing step to the following in more or less exchangeable soil fractions as also observed for other HMs after a soil washing procedure (Barona et al. 2001). This latter phenomenon can



**Fig. 1** Cu extraction efficiency at EDDS:M ratio ranging from 1 to 30 and L/S ratio equal to 15 (a). L/S ratio ranging from 15 to 45 and EDDS:M ratio equal to 10 (b)



**Fig. 2** Extracted Cu percentage related to each washing step in the four experimental setups of the SF washing configuration

cause limited increase/decrease of extracted Cu despite that low percentage value variations can be observed (Fig. 2).

Nonetheless, a general extraction trend from washing steps 1 to 8 is clearly definable for all the investigated tests. In the steps 1 and 2, A1 test showed higher efficiency than A2, A3, and A4 tests. Following washing steps did not display significant Cu extraction percentage for A1 test. In contrast, the A2, A3, and A4 tests showed long-lasting Cu extraction percentage throughout the washing steps despite low values of extracted Cu were achieved. The different behavior revealed from the four tests can be ascribable to the different operational conditions involved in terms of EDDS dose fractionation for A1, A2, and A3 tests and treatment time fractionation for A4 test. Further reason can be due to the different trend of EDDS:M ratio observed for the different tests (Table 5).

In the test A1, a sharp increase of EDDS:M ratio in the washing steps 1 and 2 can be observed, followed by a plateau from the third to the last step. This tendency is consistent with the Cu extraction results displayed in Fig. 2, where a dramatic decrease took place from the third washing step onward. Also, the EDDS:M ratio trend reported in Table 5 suggested that the washing condition involved in the A1 test entails a very fast kinetic in the first treatment hours. This makes the A1 condition suitable for rapid treatment on soil characterized by contamination that is mainly bound to non-detrital fractions.

For A2 and A3 tests, more variable EDDS:M ratio can be observed in the washing steps (Table 5) since EDDS increasing and decreasing dose was used step by step, respectively. For A2 condition, the aim was to test if better Cu extraction percentage was achievable by involving higher EDDS moles to the last washing steps where residual Cu concentration in soil detrital fractions was mainly expected. In contrast, A3 condition aimed at investigating the possibility to increase process kinetic by involving higher EDDS moles in the initial

**Table 5** EDDS:M ratio at each washing step for A1, A2, A3, and A4 tests

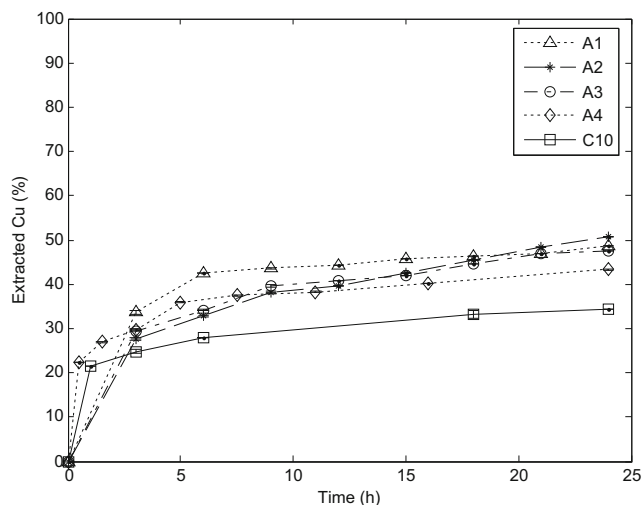
EDDS:M molar ratio (mol/mol)								
Test	Step 1	Step 2	Step 3	Step 4	Step 5	Step 6	Step 7	Step 8
A1	10.07	11.35	11.74	11.79	11.83	11.89	11.92	11.82
A2	3.14	5.62	7.94	10.35	12.06	14.36	16.73	19.16
A3	16.87	15.84	13.95	12.06	9.95	7.82	5.70	3.54
A4	10.07	10.54	10.67	10.76	10.97	11.02	11.05	11.13

washing steps and obtain faster weakly bound Cu release. In the A2 test, a linear increasing tendency for the molar ratio was observed allowing the lower but constant Cu extraction after the second washing step. In contrast, slower kinetics were observed compared to A1 test results due to the initial low dose of EDDS involved. Lower Cu extracted percentages in steps 1 and 2 than the ones achieved in A1 test were also observed for A3 test. In contrast, a higher Cu extraction in steps from 3 to 7 was observed for A3 compared to A1 tests. Nonetheless, it was also observed a process kinetic decrease corresponding to the EDDS:M ratio lowering step by step. Compared to the Cu extraction yield displayed from the A1 configuration, the ones observed for A2 and A3 suggested that these configurations are well suited for soil remediation cases where several washing steps are needed. This was indicated by the extraction efficiency enhancement of the washing steps from 3 to 8 and especially in the A2 configuration where increasing EDDS:M ratio was obtained.

A4 washing condition was implemented by decreasing the treatment time of the initial steps and by increasing the treatment time of the final steps. In this case, the aim was to extract Cu present in non-detrital soil fractions with very fast washing steps and to enhance extraction of less labile Cu forms with final longer washing steps. However, the results did not display significant improvement in terms of Cu extraction efficiency for each washing step. Treatment time decrease during the initial washing steps did not allow to achieve high Cu extraction percentage. Furthermore, extraction enhancement was not observed for steps 7 and 8 corresponding to a treatment time of 5 and 8 h, respectively. Despite the long treatment time, this could be ascribed to the lower residual Cu concentration in the soil non-detrital fractions after six washing steps. Nonetheless, comparable Cu extraction with A2 and A3 configurations in 6 h were achieved with a total treatment time of 5 h, corresponding to the washing carried out until step 4.

#### Cu extraction kinetics

Further comparison can be made between single-stage CSTR and SF configurations. Figure 3 shows the Cu cumulative extraction for the tests A1, A2, A3, and A4 and the test carried



**Fig. 3** Comparison of the cumulative extracted Cu for the tests C10, A1, A2, A3, and A4

out at EDDS:M ratio equal to 10 and L/S = 25 in the single-stage CSTR condition (C10).

Results showed that A1 condition allowed to achieve a fast Cu extraction enhancement after 3 h of total treatment time compared to extracted Cu observed in test C10. In contrast, A2 and A3 tests showed significantly higher Cu cumulative extraction than C10 test after 9 h following similar extraction tendency. Results related to A2 test also displayed slight higher extracted Cu percentage than A1 test after 21 and 24 h, while almost comparable extraction was observed for A1 and A3 test after 21 h (Fig. 3). The slight extraction enhancement obtained in A2 condition after 21 h was probably ascribable to the increasing EDDS:M ratio involved in the test achieving Cu extraction from less exchangeable soil fraction and/or lower main cation competition phenomenon effect. Furthermore, results observed for A1, A2, and A3 experimental conditions were consistent with previous studies reporting that multi-step washing configuration can improve the HM extraction efficiency of the soil washing process. In particular, multi-step washing configuration allows to involve lower chelating agent dose compared to single-step washing (Finzgar and Lestan 2007; Hong et al. 2008; Mohanty and Mahindrakar 2011), and also, further improvement are represented by shorter treatment time needed to achieve comparable HM extractions (Theodoratos et al. 2000). This was confirmed by results related to the comparison between C10 and A4 tests (Fig. 3), showing that extracted Cu was higher after 1.5 h in test A4 than after 1 h in test C10. Then, cumulative extracted Cu from A4 test remained higher than C10 values all along the test. Nonetheless, overall extracted Cu achieved in A4 was considerably lower than the extraction observed in tests A1, A2, and A3, despite the longer treatment time involved in the A4 steps 7 and 8. This further demonstrated the higher effect of the EDDS dose fractionation than the treatment time on the soil washing process efficiency. Moreover, a

comparison in terms of process kinetics was carried out among C10, A1, A2, A3, and A4 tests. First-order kinetics were used to describe the Cu extraction process. The first-order equation used for the specific case is the following:

$$\frac{d[\text{Cu}]}{dt} = -k \cdot t \tag{1}$$

where [Cu] is the Cu concentration (mg kg<sup>-1</sup>), *t* is the treatment time (h), and *k* is the first-order kinetic constant (h<sup>-1</sup>). Integrating Eq. (1) results in the following:

$$[\text{Cu}] = [\text{Cu}]_0 \cdot e^{-k \cdot t} \tag{2}$$

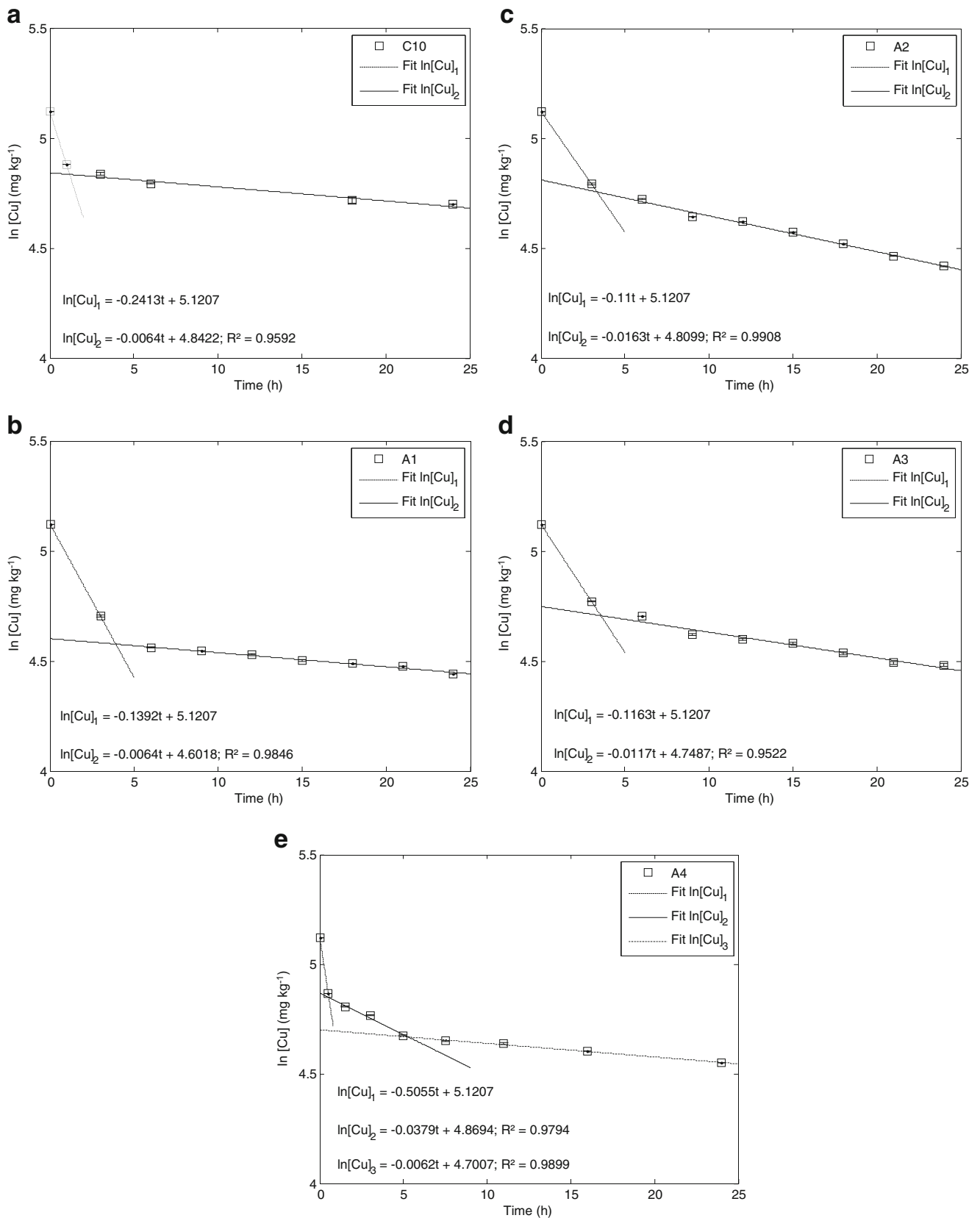
where [Cu]<sub>0</sub> is the initial Cu concentration in the soil. Finally, Eq. (2) can be modified in a linear form in order to determine *k*:

$$\ln[\text{Cu}] = \ln[\text{Cu}]_0 - k \cdot t \tag{3}$$

Figure 4a–e shows the plot of ln[Cu] decrease in the soil versus *t* for tests C10, A1, A2, A3, and A4, respectively. Moreover, the values of Eq. (3) terms determined for each test are also reported (Fig. 4a–e). One could notice that the experimental data follow an exponential feature that can be approximately divided in two kinetic steps for C10, A1, A2, and A3 tests (Fig. 4a–d) and three kinetic steps for A4 test (Fig. 4e). The two kinetic steps were distinguished with ln[Cu]<sub>1</sub> and ln[Cu]<sub>2</sub> for the first and second kinetic steps, respectively, while *k*<sub>1</sub> and *k*<sub>2</sub> represented their related kinetic constants. The term ln[Cu]<sub>3</sub> was used for the third kinetic step, and *k*<sub>3</sub> was its related kinetic constant. In particular, results from C10, A1, A2, and A3 tests showed an initial faster kinetic step followed by a slower one until end of the washing process. Initial fast kinetic step was occurring up to 1 h of treatment for C10 and 3 h of treatment for A1, A2, and A3 configurations. This assumption was strengthened by Cu cumulative extraction observed in Fig. 3, reporting a 62.3% of the total Cu extraction occurring after 1 h for C10 and 69.1, 54.1, and 62.1% of the total Cu extraction occurring after 3 h for A1, A2, and A3, respectively. Similarly for test A4, first kinetic step was limited to the initial treatment time corresponding to 0.5 h, after which 51.6% of the total Cu cumulative extraction were observed (Fig. 3). Furthermore, A4 test showed a second kinetic step occurring between 1.5 and 5 h of treatment, where 31.4% of total Cu extraction were achieved. Only 17% of total Cu extraction were observed for the last stage of the washing process corresponding to a third slowest kinetic step.

According to the cumulative extracted Cu percentages, it can be observed that initial faster kinetics are obtained for C10, A1, and A4 configurations (Fig. 3). In particular, C10 results displayed faster initial kinetics than A1 configuration due to similar extracted Cu percentage achieved in less treatment time (1 h). In contrast, similar initial kinetics were



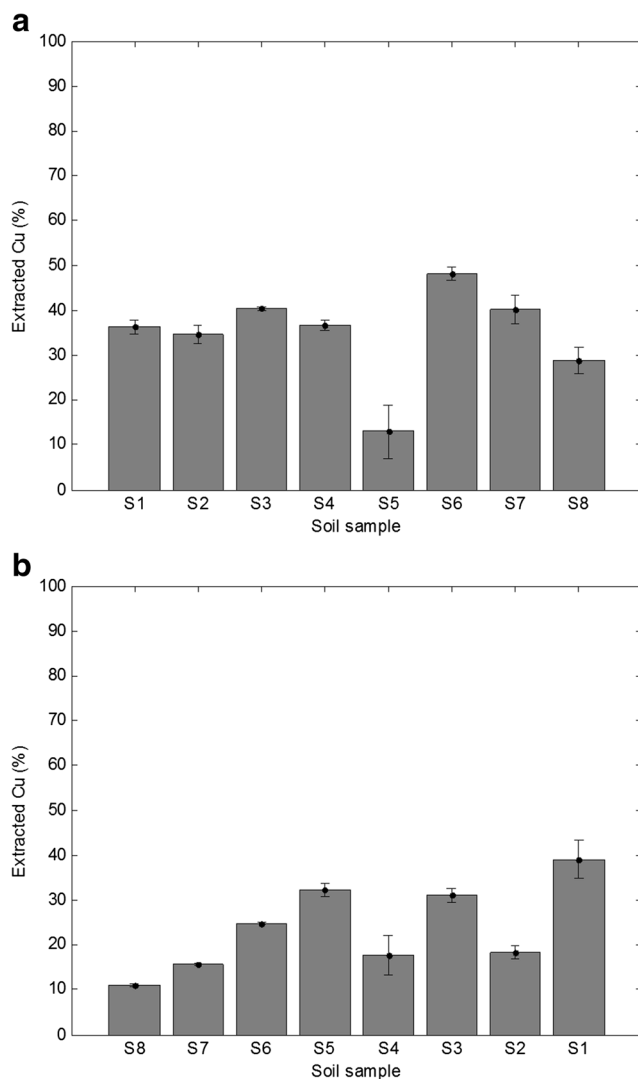


**Fig. 4** Cu concentration decrease in soil and first-order kinetic equation fitting with two kinetic steps (Fit  $\ln[\text{Cu}]_1$  and Fit  $\ln[\text{Cu}]_2$ ) for C10 test (a), A1 test (b), A2 test (c), and A3 test (d) and three kinetic steps (Fit  $\ln[\text{Cu}]_1$ , Fit  $\ln[\text{Cu}]_2$ , and Fit  $\ln[\text{Cu}]_3$ ) for A4 test (e)

observed comparing C10 and A4 configurations. However, comparable kinetics were observed for C10 and A1 second steps and A4 third step ( $k_2 = 0.0064 \text{ h}^{-1}$  for C10 and A1,  $k_3 = 0.0062 \text{ h}^{-1}$  for A4). Higher kinetic constant values ( $k_2 = 0.0163 \text{ h}^{-1}$  for A2 and  $k_2 = 0.0117 \text{ h}^{-1}$  for A3) were obtained for A2 and A3 tests in the second kinetic step compared to C10 and A1 configurations, suggesting best suitability of A2 and A3 treatments for long-lasting washings. Finally, comparison of  $k_2$  values among A2, A3, and A4 tests displayed higher values of  $k_2$  for A4 than A2 and A3 configurations ( $k_2 = 0.0163 \text{ h}^{-1}$  for A2,  $k_2 = 0.0117 \text{ h}^{-1}$  for A3, and  $k_2 = 0.0379 \text{ h}^{-1}$  for A4). This is in accordance to the comparable extracted Cu percentage achieved in lower treatment time with A4 test (5 h) than A2 and A3 tests (6 h).

**Counter-current washing configuration**

Figure 5a-b shows the results related to the CC washing configuration in terms of Cu extraction efficiency related to each step. Washing steps from S1 to S8 samples showed an almost similar Cu extraction yield for each test (Fig. 5a). The highest extraction kinetic drop is observed for the S5 sample. Similar results were observed in a previous study where an electrochemically recovered EDDS solution was involved in a multi-step soil washing (Ferraro et al. 2015). Besides the possible occurrence of the regenerated EDDS solution degradation, competition phenomena between cation competitors and Cu for EDDS chelation is a possible reason for process kinetic slowdown after five washing steps (Ferraro et al. 2015). Extracted Cu percentage was then followed by a sharp increase on the S6 sample and a slighter decrease in the following steps (Fig. 5a). The higher Cu extraction achieved for S6, S7, and S8 samples after the extraction drop observed for S5 is reasonable, considering that competition phenomena can occur and affect the process efficiency at various extent according to different process conditions. It was reported that high influence on EDDS-enhanced washing performance is displayed by cation competitors (i.e., Ca and Fe) especially in acidic conditions (Vandevivere et al. 2001). Alkaline pH observed in the present study could be a possible reason for limited competition phenomena displayed by the overall Cu extraction yield in the first washing phase. Further reason could derive from the less stable complexes that cation competitors, such as Ca, form with chelating agent compared to contaminant HMs (Papassopi et al. 1999). This can entail the detachment of the previously formed cation competitor-chelant complex leading to the further interaction between HMs and newly fed chelant molecule. The previously mentioned aspects can represent favorable factors that play a crucial role for the soil washing efficiency. For instance, a positive effect can be represented by limited influence of competition phenomena for chelation on process efficiency as also observed in Fig. 5a.



**Fig. 5** Extracted Cu percentage in the counter-current washing configuration for the first washing phase (a) and second washing phase (b)

From Fig. 5b, it was possible to observe that fresh EDDS solution addition for further soil treatment did not initially display significant Cu extraction while higher values are achieved in following steps. Moreover, generally lower extractions were observed for all the steps in the second washing compared to first washing phase. This was ascribable to the possible Cu reduction in the non-detrital soil fractions during first washing phase while high Cu amount in the detrital fractions was left during the second washing phase. As a consequence, lower Cu extraction extent was achieved during the second washing phase probably due to the presence of main residual Cu bound to the soil detrital fractions (oxidizable and residual fractions).

More clear process kinetics were observed from results displayed in Fig. 5b. Cu extraction is characterized by a main increasing tendency step by step besides two decreases for S4 and S2 samples. These discrepancies could be due to

circumscribed competition phenomena as previously observed for the first washing phase (Fig. 5a). Then, despite an overall extraction trend can be observable from one washing step to the following, possible variance can occur when focusing at single washing step. This is also due to different ion exchange phenomenon conditions concerning the specific washing step and/or Cu mobilization in different soil fractions after first washing phase.

An increase of about 27% of the overall cumulative extracted Cu was observed after a total treatment time of 24 h for the CC configuration compared to the C10 test of single-stage CSTR configuration (data not shown). Higher cumulative extracted Cu of 36.8% was achieved during the first washing phase after 12 h of treatment, while only 24.7% of cumulative Cu were extracted during the second washing phase (data not shown). Cumulative extracted Cu percentage in the first washing phase after 12 h was higher than the one achieved after a treatment of 24 h in C10 test (34.4%), whereas comparable cumulative extracted Cu was achieved in tests A1, A2, A3, and A4 for shorter treatment time (Fig. 3). This suggested that CC configuration enhanced the process kinetic in terms of Cu extraction only compared to the single-stage CSTR configuration.

Nonetheless, improvement of the overall process efficiency was observed comparing CC results with both the single-stage CSTR and SF results. This general process enhancement in the CC configuration could be mainly ascribable to the fractionation of total amount of soil to treat in the eight washing steps and the concomitant involvement of a fixed amount of EDDS dose in the washing solution. This led to the substantial increase of the L/S ratio avoiding main cation competitor effect due to the concurrent lowering of both HMs and EDDS moles as previously observed for single-stage CSTR tests (Fig. 1b).

### Practical implications for full-scale soil washing

The various washing conditions that were investigated in the present study can represent a valuable alternative to single-stage CSTR configuration for soil washing full-scale application. In fact, by comparing single-stage CSTR with results from SF and CC configurations, a process enhancement in terms of Cu extraction yield and process kinetics for both multi-stage washing configurations can be observed. This is mainly characterized by the achievement of higher extracted Cu percentages with lower treatment time and EDDS dose involved compared to the single-stage washing condition. These results can further lead to various benefits for the operator/company according to economic/operational criteria established for the soil washing technique.

EDDS dose lowering implies a lower amount of EDDS per kg of soil required for proper washing treatment. This leads to two main beneficial effects for soil washing. The first one is

represented by process cost decrease, especially for full-scale applications where high amount of soil is generally involved in the treatment. A second aspect is related to the possibility of an environmentally safer soil reuse after treatment due to the involvement of a reduced total EDDS amount per kg of soil.

Treatment time decrease implies additional advantages, which can be either the decrease of the full-scale reactor size and thus the plant costs or the increase of the soil amount that can be treated. In this latter case, the main benefit derives from the involvement of a faster process and the decrease of overall time required for a contaminated area remediation. This can lead to a fleeting treated soil disposal in the original place of collection and rapid remediation of the contaminated area.

### Conclusions

In the present study, a comparative investigation between single-stage CSTR and multi-washing configurations displayed interesting soil washing enhancement perspective as outcome. In particular, involvement of both alternative multi-washing configurations (SF and CC) highlighted significant process improvements in terms of Cu removal kinetics and yield compared to the classic single-stage CSTR configuration. Despite overall process enhancement results were achieved by involving all the washing conditions (A1, A2, A3, and A4) of SF configuration, data from A1 test displayed the best process kinetic enhancement with maximum extracted Cu percentage in 6 h. Then, constant EDDS dosage and treatment time involved for each A1 washing step allowed to achieve high Cu extraction percentage in less time than other tests.

In the CC configuration, higher overall Cu extraction percentage were observed compared to single-stage CSTR and SF configurations after an overall treatment time of 24 h. Nonetheless, A1 condition represented the best compromise between contaminant extraction efficiency and process kinetic enhancement, resulting in an interesting perspective for further investigation to test its cost-effectiveness for full-scale applications. Higher process efficiency at lower chelant dose and treatment time could make soil washing a feasible soil treatment option in economic terms besides operational ones. However, different reactor design and process control may be required by involving multi-washing configuration approach that could influence operating costs. This implies the needs for further studies on pilot- and full-scale application of the investigated configuration for cost-benefit analysis and additional assessment of process feasibility.

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## References

- Abumaizar RJ, Smith EH (1999) Heavy metal contaminants removal by soil washing. *J Hazard Mater* 70:71–86. doi:10.1016/S0304-3894(99)00149-1
- Ager P, Marshall WD (2003) Recycle of thermomechanical pulp filtrate after the removal of metals: a study with EDTA. *J Pulp Pap Sci* 29: 303–307
- Allen HE, Chen P-H (1993) Remediation of metal contaminated soil by EDTA incorporating electrochemical recovery of metal and EDTA. *Environ Prog* 12:284–293. doi:10.1002/ep.670120409
- ASTM D 2974-00 (2000) Standard test methods for moisture, ash and organic matter of peat and other organic soils. ASTM International, West Conshohocken
- ASTM D 422-63 (2007) Test method for particle-size analysis of soils. ASTM International, West Conshohocken
- Barona A, Aranguiz I, Elias A (2001) Metal associations in soils before and after EDTA extractive decontamination: implications for the effectiveness of further clean-up procedures. *Environ Pollut* 113: 79–85. doi:10.1016/S0269-7491(00)00158-5
- Begum ZA, Rahman IMM, Tate Y et al (2012) Remediation of toxic metal contaminated soil by washing with biodegradable aminopolycarboxylate chelants. *Chemosphere* 87:1161–1170. doi: 10.1016/j.chemosphere.2012.02.032
- Dermont G, Bergeron M, Mercier G, Richer-Lafleche M (2008) Soil washing for metal removal: a review of physical/chemical technologies and field applications. *J Hazard Mater* 152:1–31. doi:10.1016/j.jhazmat.2007.10.043
- Di Palma L, Ferrantelli P, Merli C, Biancifiore F (2003) Recovery of EDTA and metal precipitation from soil flushing solutions. *J Hazard Mater* 103:153–168. doi:10.1016/S0304-3894(03)00268-1
- Di Palma L, Ferrantelli P, Medici F (2005) Heavy metals extraction from contaminated soil: recovery of the flushing solution. *J Environ Manag* 77:205–211. doi:10.1016/j.jenvman.2005.02.018
- Elliott HA, Linn JH, Shields GA (1989) Role of Fe in extractive decontamination of Pb-polluted soils. *Hazard Waste Hazard Mater* 6:223–229. doi:10.1089/hwm.1989.6.223
- Espino-Mesa M, Arbelo CD, Hernandez-Moreno JM (1993) Predicting value of diagnostic soil properties on actual and potential cation exchange capacity (CEC) in Andisols and andic soils. *Commun Soil Sci Plant Anal* 24:2569–2584
- Fabbricino M, Ferraro A, Del Giudice G, d'Antonio L (2013) Current views on EDDS use for ex situ washing of potentially toxic metal contaminated soils. *Rev Environ Sci Bio/Technol* 12:391–398. doi: 10.1007/s11157-013-9309-z
- Ferraro A, van Hullebusch ED, Huguenot D et al (2015) Application of an electrochemical treatment for EDDS soil washing solution regeneration and reuse in a multi-step soil washing process: case of a Cu contaminated soil. *J Environ Manag* 163:62–69. doi:10.1016/j.jenvman.2015.08.004
- Ferraro A, Fabbricino M, van Hullebusch ED et al (2016a) Effect of soil/contamination characteristics and process operational conditions on aminopolycarboxylates enhanced soil washing for heavy metals removal: a review. *Rev Environ Sci Bio/Technol* 15:111–145. doi:10.1007/s11157-015-9378-2
- Ferraro A, Fabbricino M, van Hullebusch ED, Esposito G (2016b) Calibration and validation of a two-step kinetic mathematical model for predicting Cu extraction efficiency in an EDDS-enhanced soil washing. *Water Air Soil Pollut* 227:1–11. doi:10.1007/s11270-016-2764-8
- Finzgar N, Lestan D (2007) Multi-step leaching of Pb and Zn contaminated soils with EDTA. *Chemosphere* 66:824–832. doi:10.1016/j.chemosphere.2006.06.029
- Fischer K, Bipp H-P, Riemschneider P et al (1998) Utilization of biomass residues for the remediation of metal-polluted soils. *Environ Sci Technol* 32:2154–2161. doi:10.1021/es9706209
- Guo H, Wang W, Sun Y et al (2010) Ethyl lactate enhances ethylenediaminedisuccinic acid solution removal of copper from contaminated soils. *J Hazard Mater* 174:59–63. doi:10.1016/j.jhazmat.2009.09.016
- Hauser L, Tandy S, Schulin R, Nowack B (2005) Column extraction of heavy metals from soils using the biodegradable chelating agent EDDS. *Environ Sci Technol* 39:6819–6824. doi:10.1021/es050143r
- Hong PKA, Cai X, Cha Z (2008) Pressure-assisted chelation extraction of lead from contaminated soil. *Environ Pollut* 153:14–21. doi:10.1016/j.envpol.2007.12.022
- Kim C, Lee Y, Ong SK (2003) Factors affecting EDTA extraction of lead from lead-contaminated soils. *Chemosphere* 51:845–853. doi:10.1016/S0045-6535(03)00155-3
- Komárek M, Száková J, Rohošková M et al (2008) Copper contamination of vineyard soils from small wine producers: a case study from the Czech Republic. *Geoderma* 147:16–22. doi:10.1016/j.geoderma.2008.07.001
- Koopmans GF, Schenkeveld WDC, Song J et al (2008) Influence of EDDS on metal speciation in soil extracts: measurement and mechanistic multicomponent modeling. *Environ Sci Technol* 42:1123–1130. doi:10.1021/es071694f
- Lestan D, Luo C, Li X (2008) The use of chelating agents in the remediation of metal-contaminated soils: a review. *Environ Pollut* 153:3–13. doi:10.1016/j.envpol.2007.11.015
- Levenspiel O (1972) Chemical reaction engineering. J Wiley and Sons, New York
- Lim T-T, Chui P-C, Goh K-H (2005) Process evaluation for optimization of EDTA use and recovery for heavy metal removal from a contaminated soil. *Chemosphere* 58:1031–1040. doi:10.1016/j.chemosphere.2004.09.046
- Mohanty B, Mahindrakar AB (2011) Removal of heavy metal by screening followed by soil washing from contaminated soil. *Int J Technol Eng Syst* 2:290–293
- Papassiopi N, Tambouris S, Kontopoulos A (1999) Removal of heavy metals from calcareous contaminated soils by EDTA leaching. *Water Air Soil Pollut* 109:1–15. doi:10.1023/A:1005089515217
- Peters RW (1999) Chelant extraction of heavy metals from contaminated soils. *J Hazard Mater* 66:151–210. doi:10.1016/S0304-3894(99)00010-2
- Pociecha M, Lestan D (2010) Electrochemical EDTA recycling with sacrificial Al anode for remediation of Pb contaminated soil. *Environ Pollut* 158:2710–2715. doi:10.1016/j.envpol.2010.04.014
- Pueyo M, Mateu J, Rigol A et al (2008) Use of the modified BCR three-step sequential extraction procedure for the study of trace element dynamics in contaminated soils. *Environ Pollut* 152:330–341. doi: 10.1016/j.envpol.2007.06.020
- Race M, Marotta R, Fabbricino M et al (2016) Copper and zinc removal from contaminated soils through soil washing process using ethylenediaminedisuccinic acid as a chelating agent: a modeling investigation. *J Environ Chem Eng* 4:2878–2891. doi:10.1016/j.jece.2016.05.031
- Sarkar D, Andra SS, Saminathan SKM, Datta R (2008) Chelant-aided enhancement of lead mobilization in residential soils. *Environ Pollut* 156:1139–1148. doi:10.1016/j.envpol.2008.04.004
- Satyro S, Race M, Marotta R et al (2016) Photocatalytic processes assisted by artificial solar light for soil washing effluent treatment. *Environ Sci Pollut Res*. doi:10.1007/s11356-016-6431-9

- Steele MC, Pichtel J (1998) Ex-situ remediation of a metal-contaminated superfund soil using selective extractants. *J Environ Eng* 124:639–645. doi:10.1061/(ASCE)0733-9372(1998)124:7(639)
- Subirés-Muñoz JD, García-Rubio A, Vereda-Alonso C et al (2011) Feasibility study of the use of different extractant agents in the remediation of a mercury contaminated soil from Almaden. *Sep Purif Technol* 79:151–156. doi:10.1016/j.seppur.2011.01.032
- Sun B, Zhao FJ, Lombi E, McGrath SP (2001) Leaching of heavy metals from contaminated soils using EDTA. *Environ Pollut* 113:111–120. doi:10.1016/S0269-7491(00)00176-7
- Tandy S, Bossart K, Mueller R et al (2004) Extraction of heavy metals from soils using biodegradable chelating agents. *Environ Sci Technol* 38:937–944. doi:10.1021/es0348750
- Tandy S, Ammann A, Schulin R, Nowack B (2006) Biodegradation and speciation of residual SS-ethylenediaminedisuccinic acid (EDDS) in soil solution left after soil washing. *Environ Pollut* 142:191–199. doi:10.1016/j.envpol.2005.10.013
- Theodoratos P, Papassiopi N, Georgoudis T, Kontopoulos A (2000) Selective removal of lead from calcareous polluted soils using the Ca-EDTA salt. *Water Air Soil Pollut* 122:351–368. doi:10.1023/A:1005295119231
- Tsang DCW, Lo IMC, Surampalli RY (2012) Design, implementation, and economic/societal considerations of chelant-enhanced soil washing. In: Tsang DCW, Lo IMC, Surampalli RY (eds) Chelating agents for land decontamination technologies. American Society of Civil Engineers, Reston, pp 1–26
- USEPA (1995) SW-846 EPA method 3051: microwave assisted acid digestion of sediments, sludges, soils and oils, in: test methods for evaluating solid waste, 3rd Edn, 3rd Update. US Environmental Protection Agency, Washington DC
- Van Benschoten JE, Matsumoto MR, Young WH (1997) Evaluation and analysis of soil washing for seven lead-contaminated soils. *J Environ Eng* 123:217–224. doi:10.1061/(ASCE)0733-9372(1997)123:3(217)
- Vandevivere P, Hammes F, Verstraete W et al (2001) Metal decontamination of soil, sediment, and sewage sludge by means of transition metal chelant [S,S]-EDDS. *J Environ Eng* 127:802–811. doi:10.1061/(ASCE)0733-9372(2001)127:9(802)
- Voglar D, Lestan D (2010) Electrochemical separation and reuse of EDTA after extraction of Cu contaminated soil. *J Hazard Mater* 180:152–157. doi:10.1016/j.jhazmat.2010.04.007
- Zeng QR, Sauvé S, Allen HE, Hendershot WH (2005) Recycling EDTA solutions used to remediate metal-polluted soils. *Environ Pollut* 133:225–231. doi:10.1016/j.envpol.2004.06.006
- Zou Z, Qiu R, Zhang W et al (2009) The study of operating variables in soil washing with EDTA. *Environ Pollut* 157:229–236. doi:10.1016/j.envpol.2008.07.009