Optimization of parameters for the extraction of Pb from lateritic soil using EDTA

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Abstract

In this study, we optimized the factors influencing the remediation of Pb-contaminated laterite soil by Ethylenediaminetetraacetic acid (EDTA) washing technology. Soil was artificially contaminated with Pb and laboratory extractions were carried out by varying (1) pH of EDTA, (2) EDTA–Pb molar ratio, (3) Solution–soil ratio and (4) the number of steps in the sequential extraction process. The batch extraction experiments were designed using central composite design (CCD)-based response surface methodology by varying these four factors at five different levels. The range adopted for these factors were; pH (4–8), EDTA–Pb molar ratio (0–4), Solution–soil ratio (3–21) and number of steps of extraction (1–5). The significance and adequacy of the model given by CCD were tested using analysis of variance (ANOVA). Numerical optimization results showed that the maximum extraction efficiency of 90.82% is obtained at a desirability of 0.97 when pH of EDTA was 4, EDTA–Pb molar ratio was 3, solution–soil ratio was 16.5 and the number of steps was 4. Laboratory extraction carried out adopting the numerically optimized factors gave an extraction efficiency of 89.6%. The optimized parameters can be adopted when ex situ remediation of Pb-contaminated laterite soil is carried out using EDTA washing technique.

Keywords Pb extraction · Laterite soil · Chelating agent · EDTA · Central composite design · Response surface methodology

1 Introduction

Heavy metal contamination of soil is a concern because of the non-degradable nature of the contaminant. In situ methods for remediating heavy metal-contaminated soil involves either extracting the contaminants from soil or its immobilization in the contaminated soil. Risk reduction happens, in the first case because the soil is rendered non-hazardous and in the second case because the chances of exposure to the contaminants are significantly reduced. Ex situ remediation methods also follow similar principles. Heavy metals are extracted from the soil at a facility installed for the purpose and the treated soil may be put back at the site. Alternately, the contaminated soil may be dumped in a landfill where they are sealed from contact with the outside environment.

Soil washing is an ex situ method suitable for extracting heavy metals from contaminated soil. The heavy metals are solubilized in a washing solution and removed from the soil [1–10]. Among the different agents for soil washing, acids and chelating agents are the most powerful extractants [10–16]. But there are chances that acidic solutions cause deteriorations of the soil structure. Thus, chelating agents as soil washing fluid can be considered as an effective and less disruptive method of decontamination [9, 17, 18]. Among the variety of chelating agents, EDTA is the most commonly used and powerful one for extraction due to its efficiency, availability, recoverability and relatively low cost. A good number of works were reported on the extraction of heavy metals from contaminated soil using EDTA.

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Extraction of heavy metals using EDTA depends on a number of factors such as pH of EDTA [19, 20], concentration of EDTA [19–23], liquid–solid ratio [24] and the reaction time [20, 21, 24]. The general trend was that the extraction efficiency increased with increasing EDTA–metal ratio and liquid–solid ratio. The extraction was efficient at acidic pH and the reaction time beyond a limit was not improving the extraction efficiency.

Lead (Pb) is a heavy metal contaminant frequently detected in the soil environment all around the world [25–30], especially in urban centres [31, 32]. The maximum tolerable soil concentration of Pb based on human health protection is 84 mg/kg [33]. Pb is a probable human carcinogen (Type B2) as per USEPA weight of Evidence classification [34] and health effects associated with exposure include neurotoxicity, developmental delays, hypertension, impaired hearing acuity, impaired haemoglobin synthesis and male reproductive impairment [34]. The affinity of Pb for sorption onto soil is higher when compared to many other heavy metals [35]. This can result in a higher Pb concentration build-up in the soil and consequently higher Pb exposure to people coming in contact with the soil, directly or indirectly.

Pb-contaminated soil can be remediated by washing using EDTA. As in the case of most heavy metals, the extraction efficiency of Pb from soil using EDTA depends on factors like solution: soil ratio, major cations present in soils and the EDTA: Pb molar ratio. Kim et al. [36] observed that lead extraction efficiencies for soils were different for different stoichiometric ratios; but if a sufficiently large amount of EDTA was applied, most of the lead was extracted from the soils. The different extraction efficiencies for the same [EDTA] to [Pb] molar ratio were explained by the presence of other heavy metals that compete with Pb for combining with EDTA. Elliot and brown [37] in their studies observed that 95% of lead present in the soil was removed when an EDTA–Pb molar ratio of 2:1 was adopted. At equimolar ratio, the removal rate was very less. Removal of heavy metals was very high at lower pH, acidic media [38]. These and many other studies have shown that a contact time of approximately 2 h [24, 39, 40], an [EDTA]: [Pb] molar ratio of about 2:1 [39] and an acidic pH [24, 37, 41] are ideal for the extraction of Pb from soil using EDTA.

Though the extraction of Pb from soil using EDTA is a topic that has been widely studied in different parts of the world from as early as late 1970's [42], the research literature is notably scarce on studies conducted with laterite soil. Laterite soils are rich in iron and aluminium, low in silica, chemically acidic, and exhibit a soil profile different from that of other soils [43]. They are derived from a wide variety of rocks weathering under strongly oxidizing and leaching conditions. It is formed in tropical and

subtropical regions where the climate is humid. The soil type is important because of its significant geographical coverage [43]. Thus, in spite of the rich literature available on the extraction of Pb using EDTA, a study of the factors influencing the extraction of Pb from laterite soil using EDTA is relevant.

Another factor that is not studied extensively is the number of steps adopted for extraction retaining the overall solution–soil ratio constant. For the same EDTA–Pb molar ratio, one can carry out the extraction in more than one step, using a fraction of the total required EDTA each time. Though, this may increase the effort, it is worth investigating such stepwise extraction as there is a potential for higher Pb removal for the same amount of EDTA used. Also, the use of an efficient method to design the experimental programme to get reliable results from the experimental runs was missing in most studies.

Therefore, the objectives of the reported study were (1) investigating the factors that affects the extraction of lead using EDTA from contaminated laterite soil (2) applying central composite design (CCD)-based response surface methodology (RSM) to analyse the effects of different factors on extraction efficiency and to get the optimum values of the factors for attaining a maximum efficiency.

2 Materials and methods

2.1 Uncontaminated lateritic soil

The lateritic soil used in the study was collected from the National Institute of Technology, Calicut premises, where the study was carried out. The soil type in the region is laterite. The surface soil was collected from a depth of 10–15 cm and kept within polythene bags to preserve the moisture content. Then, the soil was oven dried. Table 1 shows the physical and geochemical characteristics of the selected soil sample and the corresponding methods used for measurement. Indian Standard (IS) procedures were adopted wherever available. From the oven-dried sample, soil with particle size less than 2 mm was taken for further studies. Particles of larger size tend to be less reactive in short term due to their higher mass to surface area ratio. Hence the larger particles were avoided in the experimental study.

2.2 Soil contamination

The soil sample was spiked with lead nitrate solution such that the target concentration of lead was about 1000 mg/kg. It is then placed in an orbital incubating shaker at a speed of 180 rpm at room temperature for 48 h [44]. This is the wet ageing period and is necessary for complete

Table 1 Characteristics of the uncontaminated soil

Soil property	Value	Method/equipment used	Reference code/standard
Soil pH	5.98	pH metre	ASTM D4972
Electrical conductivity (Ms)	0.45	Conductivity metre	IS 14767:2000
Natural moisture content (%)	19.75	Oven drying method	IS 2720 (PART-2): 1973
Specific gravity	2.33	Pycnometer method	IS 2720 (PART-3): 1980
Total organic carbon (ppm)	51.21	TOC analyser	US EPA Method 9060A
Total organic matter (%)	0.69	Loss on ignition (LOI) method Muffle furnace	ASTM D2974-00
Cation exchange capacity (meq/100 g)	43.25		IS 2720 (PART-24): 1976
Particle size			
Clay (%)	6.25	Standard IS sieves (Sieve no. 4.75 mm, 2.36 mm, 1.18 mm, 600 μ , 300 μ , 150 μ and 75 μ)	IS 2720 (PART-4): 1985
Silt (%)	2.28		
Sand (%)	77.21		
Gravel (%)	22.79		

subjection of every soil particle to contamination. At the end of the 48-h wet ageing period, the suspension was centrifuged to separate soil and solution. Distilled water was used for washing the contaminated soil to remove the entrapped water from the soil. Then, it was dried and aged further for another 6 weeks as dry aging.

2.3 Acid digestion

For finding out the initial concentration of lead in the artificially contaminated soil, the soil was subjected to acid digestion in order to dissolve the metal salts. One gram of soil sample was treated with 50 mL Aqua Regia and 10 mL 1:1 H_2SO_4 . The mixture was dried on a hot plate, and then, it was cooled to room temperature (37 °C). This digested soil sample was then boiled with 10 mL Hydrochloric acid and 1 mL concentrated HNO_3 and 50 mL water to re-dissolve the metal salts. The extract was then filtered with filter papers and the volume was adjusted to 50 mL (EPA Method 3050b). Then, the solution was analysed using a Perkin Elmer PinAAcle 900 F Atomic Absorption Spectrophotometer (flame only) equipped with AA Win lab software.

2.4 Experiment design

The extraction experiments were designed using the response surface methodology (RSM). RSM allows the study of a number of independent factors influencing a process and the interactions between the factors with minimum number of experimental runs [45]. An important characteristic of RSM is the design of experiments (DOE) [46]. DOE will give the points where the output/response should be evaluated. Most commonly used method for DOE is central composite designs (CCD). A second-order

model can be constructed efficiently with CCD [47]. CCD is composed of first-order designs along with additional central and axial points which are at equal distance from the central point for estimating the parameters of the second order model. In this study, a central composite design having four independent variables, namely, pH of EDTA, EDTA–Pb molar ratio, soil–solution ratio and number of steps for extraction was considered. The efficiency of extraction (%) was considered as the dependent variable. Based on the literature studied, the range of the operating parameters was fixed. The variables and their levels are represented in Table 2. CCD was done by Design Expert Software version 10 [48].

2.5 Extraction procedure

According to the runs obtained from design of experiment using central composite design-based response surface methodology, extraction of lead from the artificially contaminated soil was carried out using EDTA solution. That is, batch extraction experiments were conducted using EDTA solution at various pH of the washing solution, different liquid/solid ratios and also by varying lead–EDTA molar ratios. The extraction solution was prepared from

Table 2 Experimental range and levels of the independent variables

Variables	Range and levels				
	– α	–1	0	+1	+ α
A: pH of EDTA	4	5	6	7	8
B: EDTA–Pb molar ratio	0	1	2	3	4
C: Solution–Soil ratio	3	7.5	12	16.5	21
D: No. of steps	1	2	3	4	5

the reagent-grade disodium ethylenediaminetetracetate ($\text{Na}_2\text{-EDTA}$) according to the requirements. The contact time for metal extraction was decided to be 2 h. The required amount of soil sample was taken and mixed with EDTA solution in a conical flask. Then, it was placed in an orbital incubating shaker operated at 180 rpm at room temperature for 2 h [24, 39]. After mixing, the sample is allowed to settle for 15 min in centrifuge tubes and then centrifuged and filtered through a Whatman no. 42 filter paper. The filtered liquid was analysed for lead concentration using Perkins-Elmer Atomic Absorption Spectrophotometer.

3 Results

3.1 Design of experiment and extraction efficiencies

A total of 30 experiments were carried out with the different sets of values of the independent variables obtained through DOE. The details of the experimental runs and the extraction efficiencies obtained in those runs are tabulated in Table 3. In these 30 runs, maximum extraction efficiency of 89.3% was obtained when the pH of EDTA was 7, EDTA–Pb molar ratio was 1, Solution–soil ratio was 16.5 and the number of steps of extraction was 4 (Run no. 25, in Table 3).

3.2 Analysis of variance (ANOVA)

Data were graphically analysed using analysis of variance (ANOVA) in order to obtain the interaction between independent variables and the responses. ANOVA is required to test the significance and adequacy of the model. The fit of the model can also be checked by the coefficient of determination, R^2 . ANOVA table is represented in Table 4.

From the ANOVA table, it is clear that the Fisher F test probability P value is relatively low. Thus, ANOVA of the quadratic regression model demonstrates that the model is significant. Here, the R^2 value which was 0.9777 tells that 97.77% variations in the extraction efficiency could be explained by the model.

The factors having $P < 0.005$ are considered significant and directly influencing the reactions. P value indicates the probability that the response occurred randomly without the influence of the independent factors. It can be seen that the factors A, B, C, D, AB, AC and AD are significant model terms. Values greater than 0.1000 indicate the model terms are not significant. So, the factors BC, BD and CD are non-significant. The empirical regression equation for extraction efficiency ($E\%$) in terms of coded factors is given in Eq. (1)

Table 3 Experimental conditions and results of central composite design

Run	pH	EDTA–Pb molar ratio	Solution–soil ratio	Total no. of steps	Extraction efficiency (%)
1	7	1	16.5	2	66.2
2	4	2	12	3	79.3
3	5	1	16.5	2	62.6
4	5	1	7.5	2	58.5
5	6	2	12	3	68
6	7	1	7.5	2	47.6
7	8	2	12	3	57.3
8	6	2	12	5	86.6
9	7	1	7.5	4	64.6
10	7	3	16.5	2	69.3
11	6	2	12	3	68
12	6	2	12	1	54
13	6	2	12	3	68
14	7	3	7.5	2	46.4
15	6	2	21	3	86
16	5	3	16.5	4	88.2
17	7	3	16.5	4	85.7
18	5	1	16.5	4	75.4
19	6	2	12	3	68
20	5	1	7.5	4	70.3
21	5	3	7.5	4	79.6
22	5	3	7.5	2	71.4
23	6	0	12	3	58.4
24	6	2	12	3	68
25	7	1	16.5	4	89.3
26	6	4	12	3	78.6
27	7	3	7.5	4	64
28	5	3	16.5	2	78.3
29	6	2	12	3	68
30	6	2	3	3	54.8

$$E = 69.35 - 3.97A + 3.7B + 7.29C + 7.58D - 3.31AB + 3.95AC + 1.96AD + 0.48BC - 0.79BD + 0.48CD \quad (1)$$

The extraction efficiencies can be predicted with the help of Eq. (1). The plot of actual versus predicted extraction efficiency for the study is illustrated in Fig. 1, which shows that these are in range with each other.

3.3 Effect of operating parameters

3.3.1 Effect of pH

Figure 2 shows the effect of pH of EDTA solution on the extraction efficiency of lead based on the batch extraction studies conducted at fixed values of EDTA–lead molar

Table 4 ANOVA results

Source	Sum of squares	df	Mean square	F value	P value Prob>F
Model	3866.36	10	386.64	83.45	< 0.0001
A	377.63	1	377.63	81.50	< 0.0001
B	328.56	1	328.56	70.91	< 0.0001
C	1276.04	1	1276.04	275.41	< 0.0001
D	1380.17	1	1380.17	297.88	< 0.0001
AB	175.56	1	175.56	37.89	< 0.0001
AC	249.64	1	249.64	53.88	< 0.0001
AD	61.62	1	61.62	13.30	0.0017
BC	3.61	1	3.61	0.78	0.3884
BD	9.92	1	4.63	2.14	0.1597
CD	3.61	1	6.29	0.78	0.3884
Residual	88.03	19	0.000		
Lack of fit	88.03	4			
Pure error	0.000	5			
Cor total	3954.39	29

Fig. 1 Predicted versus actual extraction efficiencies

ratio, solution–soil ratio and the number of steps of extraction of 3, 16.5 and 4, respectively. From the graph, it is clear that pH has an inhibitory effect on the extraction process of lead; as pH of EDTA solution increased, the efficiency of extraction decreased. This may be because, as the pH becomes more alkaline the ability of chelating agents to enhance lead solubility is decreased. At higher pHs, hydrolysis of metal with hydroxide ions may be favoured over the complexation process with EDTA.

3.3.2 Effect of EDTA–Lead molar ratio

Figure 3 shows the effect of EDTA–lead molar ratio on the extraction efficiency of lead based on the batch

Fig. 2 Effect of pH at EDTA–lead molar ratio=3, soil–solution ratio=16.5 and number of steps=4 **Fig. 3** Effect of EDTA–lead molar ratio at pH of EDTA solution=4, soil–solution ratio=16.5 and number of steps=4

extraction studies conducted at fixed values of pH of EDTA solution, solution–soil ratio and the number of steps of extraction of 4, 16.5 and 4, respectively. This factor has a positive impact on the extraction efficiency. As the molar ratio increased, the amount of lead extracted also increased. Since EDTA is a hexadentate ligand [49] which can form a very stable coordination compound with lead which has a coordination number of six, one Pb needs one EDTA molecule to form a Pb–EDTA coordination compound [36]. Since EDTA is a non-specific chelating agent, it may form compounds with other metal ions. So, more EDTA may be required to extract the heavy

metal than the amount of lead in the soil. That is, more EDTA–lead ratio will give more extraction efficiency.

3.3.3 Effect of solution–soil ratio

Figure 4 shows the effect of solution–soil ratio on the extraction efficiency of lead based on the batch extraction studies conducted at fixed values of pH of EDTA solution, EDTA–lead molar ratio and the number of steps of extraction of 4, 3 and 4, respectively. As the solution–soil ratio increased the extraction efficiency also got increased. This indicates that if the amount of EDTA is increased, the rate of extraction is also increased. But the problem with higher solution–soil ratio is that disposal of large amounts of waste water may create issues.

3.3.4 Effect of number of steps of extraction

Figure 5 shows the effect of number of steps of extraction on the extraction efficiency of lead based on the batch extraction studies conducted at fixed values of pH of EDTA solution, EDTA–lead molar ratio and the solution–soil ratio of 4, 3 and 16.5, respectively. From the graph, it could be concluded that adding EDTA solution as a whole to the soil at a time and continuing the extraction is less efficient than adding the whole amount of EDTA solution step by step and then extracting the heavy metal. So, this factor, number of steps of extraction is having a positive impact on the extraction efficiency.

3.3.5 Effect of pH of EDTA solution and EDTA–lead molar ratio

Figure 6 shows the combined effect of pH of EDTA solution and EDTA–lead molar ratio on extraction efficiency

Fig. 4 Effect of soil–solution ratio at pH of EDTA solution = 4, EDTA–lead molar ratio = 3 and number of steps = 4

Fig. 5 Effect of number of steps at pH of EDTA solution = 4, EDTA–lead molar ratio = 3 and soil–solution ratio = 16.5

under the operating conditions of solution–soil ratio = 16.5 and number of steps of extraction = 4. From the figure, it can be concluded that the higher values of the EDTA–lead molar ratio towards minimum pH give higher values of extraction efficiency than the lower values as more EDTA molecules are available for extraction of lead from the contaminated soil. Lead is more mobile in acidic medium [50]. Filipek and Pawlowski [51] showed that at low pH values, extractability increased considerably as a result of a decrease in organic and mineral colloid binding forces which in turn promoted metal leaching through the soil without the addition of EDTA.

3.3.6 Effect of pH of EDTA solution and solution–soil ratio

In order to study the effect of the interaction of pH and solution–soil ratio in extraction efficiency in lead

Fig. 6 Effect of pH and EDTA–lead molar ratio at solution–soil ratio = 16.5 and number of steps = 4

Fig. 7 Effect of pH and solution–soil ratio at EDTA–lead molar ratio=3 and number of steps=4

Fig. 8 Effect of pH and number of steps at EDTA–lead molar ratio=3 and solution–soil ratio=16.5

contaminated soils, it was varied between 0 and 3. Figure 7 shows the combined influence of pH of EDTA solution and solution–soil ratio on the lead extraction process under the conditions of EDTA–lead molar ratio of 3 and number of steps of extraction of 4. From the results, it is found that increasing the solution–soil ratio leads to increase in extraction efficiency. At lower pH, the extraction efficiency is high, because in acidic medium EDTA–lead complex is more stable.

Table 5 Goal set for each factor

Factors	Goal	Lower limit	Upper limit
pH of EDTA	In range	4	8
EDTA–Pb molar ratio	In range	1	3
Solution–soil ratio	In range	7.5	16.5
Number of steps	In range	2	4
Extraction efficiency	Maximize	60	100

Table 6 Numerical optimization for central composite design

S. no.	A	B	C	D	Response
1	4.002	3.000	16.500	4.000	90.814
2	4.017	3.000	16.480	4.000	90.793
3	4.036	3.000	16.500	4.000	90.768
4	4.000	3.000	15.740	4.000	90.760
5	4.000	3.000	15.914	3.994	90.753
6	4.000	3.000	15.452	4.000	90.738
7	4.000	3.000	16.451	3.976	90.735
8	4.028	2.998	16.500	3.993	90.732
9	4.012	3.000	15.695	4.000	90.731
10	4.000	3.000	15.164	4.000	90.715

3.3.7 Effect of pH of EDTA solution and number of steps of extraction

Figure 8 shows the combined effect of pH of EDTA solution and number of steps of extraction on extraction efficiency under the operating conditions of EDTA–lead molar ratio of 2 and solution–soil ratio of 16.5. From the figure, it can be said that the higher values of the steps of extraction towards lower pH give better extraction efficiency than the higher pH values.

3.4 Process optimization

In numerical optimization, goals should be set for every factor as well as for the response. Then, the goals are combined into an overall desirability function. Desirability is an objective function which is having a value of zero outside of the limits to a value of one at the goal. All four variables and extraction efficiency were set in the experimental ranges for maximum desirability as shown in Table 5.

Optimization results clearly showed that the desirable stretching conditions shifted to lower pH of EDTA solution

Table 7 Comparison of optimized extraction efficiency obtained from mathematical design with that obtained from experimental study

	Modelled	Experimental
Parameters		
pH of EDTA solution	4.002	4
EDTA–lead molar ratio	3.000	3
Solution–soil ratio	16.500	16.5
Number of steps	4.000	4
Extraction efficiency (%)	90.814	89.6
Error	1.34%	

and higher EDTA–Pb molar ratio, solution–soil ratio and number of steps. Table 6 shows the numerical optimization for central composite design.

This result was experimentally validated by doing the extraction of lead from the soil when pH of EDTA solution was 4 and EDTA–Pb molar ratio was 3, solution–soil ratio was 16.5 and number of steps of extraction was 4. The result is shown in Table 7.

4 Discussion

This study on the extraction of Pb using the chelating agent EDTA, from an artificially contaminated laterite soil showed that the factors considered, i.e., pH of EDTA, EDTA–Pb molar ratio, solution–soil ratio and the number of steps in the sequential extraction process, were highly influential in the extraction efficiency. These factors could explain almost 98% of the variations noticed in the extraction efficiency. The study showed that lower pHs of EDTA are preferred for the extraction process which is in agreement with the findings of previous reported studies [20, 52–55]. This may be because, as the pH become more alkaline the ability of chelating agent to enhance lead solubility is decreased, since in higher pH hydrolysis of metal with hydroxide ions may be favoured over the complexation process with EDTA.

The study showed that EDTA–Pb molar ratio has a positive impact on the extraction of lead. Similar results were reported by Naghipour et al. [53], Zou et al. [20], Kim et al. [36]. Since each Pb needs only one EDTA molecule to form EDTA–Pb coordination complex, the EDTA–Pb molar ratio is 1, stoichiometrically. But, as EDTA is a non-specific chelating agent, it may form complexes with other metals present in the soil as well, resulting in the requirement of higher EDTA–Pb molar ratio for better Pb removals. Sun et al. [40] noticed the ratios of Zn, Cd, Cu and Pb in the extract similar to those in the soils, in their batch extraction studies, suggesting that EDTA extracted

the four heavy metals with similar efficiency. The optimum EDTA–Pb molar ratio of 3 noticed in the study was in fact the result of an experiment design decision. Of the various EDTA–Pb molar ratios adopted in the study, the highest gave the best result. A higher ratio would have, perhaps, given a better extraction efficiency, but would result in higher amounts of EDTA consumption. Moreover, with EDTA–Pb molar ratio of 3, a theoretical extraction efficiency of 90.8 and an experimental extraction efficiency of 89.6 were noticed that were significant. The value of 3, in spite of the addition of Pb alone as a contaminant is explained by the presence of Iron (Fe) in laterite soil which would compete with Pb for complexation.

The optimum solution–soil ratio obtained in our study was 16.5. The general trend was, better removal for higher solution–soil ratios. Although we noticed this trend, there are studies that report more or less same extraction efficiency for a wider range of solution soil ratios. Kim et al. [36] reported that the extraction of lead from the lead-contaminated soils was not affected by a solution: soil ratio as low as 3:1 but instead was dependent on the quantity of EDTA present. Palma and Ferrantelli [56] found mixing time influencing the optimum solution–soil ratio, but, for another metal, Copper. They got same extraction efficiencies for the solution–soil ratio and mixing time combinations of 5 and 23 h and 12.5 and 5 h. In this study, mixing time was kept constant as adopted in some of the previous studies [39].

The optimum number of steps of extraction obtained was 4. Many studies have attempted the stepwise sequential extraction of Pb from soil using EDTA [20, 57]. But, an optimization of this factor was not noticed in the literature. In the optimized four-step extraction process retaining the overall solution–soil ratio constant, each step had a solution–soil ratio of 4.1, which is higher than the value of 3 reported by Kim et al. [36] as limiting.

The extraction efficiency of Pb from soil using EDTA depends on the type of soil also [36]. One reason for this is the natural presence of some metals in the soil which may compete with Pb for complexing with EDTA. The grain size, mineral compositions, etc. of soil affects its ability for the sorption of heavy metals [58, 59] and consequently their extraction using EDTA. Therefore, the factors influencing extraction need to be optimized separately for each type of soil. Our study was addressing the laterite soil. But, even within the laterite soil, the chemical and mineralogical characteristics vary across different sites and locations according to the parent material [60–62]. In this sense, a single lateritic soil type from one location might be inadequate to model all the extensive chemical behaviours of all the lateritic soil samples (contaminated with Pb) with EDTA. In spite of this limitation, this study provides a

framework for the future studies on extraction of Pb from other lateritic soil types.

5 Conclusion

In this study, artificially contaminated lateritic soil was used to study the influence of selected parameters on the efficiency of decontaminating the soil using EDTA washing. The contaminant studied was Pb. An acidic pH for the washing solution was found to give better extraction efficiency. EDTA–Pb molar ratio, solution–soil ratio and the number of steps in the sequential extraction process were positively correlated to the extraction efficiency. The regression equation developed could explain 98% of the variations in extraction efficiency. The study showed that the extraction process in multiple steps, keeping the total volume of EDTA solution the same, was giving better efficiency than single step extraction. The presence of Fe as a competing metal resulted in the requirement of EDTA at three times the stoichiometric quantities for a 90% extraction efficiency of Pb. The use of central composite design-based surface response methodology made the design and analysis of the experiment easy. Though the study was carried out on contaminated laterite soil, the results shall be extended to lateritic soils from other locations with caution as the mineralogical characteristics of the soils can vary from place to place, at times resulting in a different sets of optimum values.

Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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