Agricultural Sciences, 2019, 10, 882-892
http://www.scirp.org/journal/as
ISSN Online: 2156-8561
ISSN Print: 2156-8553

Effective Sodium Metabisulfite ($Na_2S_2O_5$), HCl, Sulfur and Distilled Water for the Removal of Pb, Zn and Cr Contaminated Soil in the Columns Method

Abdulbaset Egrira Salama

Department of Soil and Water, Faculty of Agriculture, Bani Waleed University, Bani Waleed, Libya
Email: apbo1977@gmail.com

How to cite this paper: Salama, A.E. (2019) Effective Sodium Metabisulfite ($Na_2S_2O_5$), HCl, Sulfur and Distilled Water for the Removal of Pb, Zn and Cr Contaminated Soil in the Columns Method. *Agricultural Sciences*, **10**, 882-892.
https://doi.org/10.4236/as.2019.107067

Received: March 28, 2019
Accepted: July 8, 2019
Published: July 11, 2019

Abstract

This paper presents an evaluation of different dose of Sodium Metabisulfite (0.01 M $Na_2S_2O_5$), ($Na_2S_2O_5$) + (0.1 HCl), and Distilled water for the removal of soil contaminated with Pb, Zn and Cr by the column mode. The field soil contained concentrations of Pb (307.31 $mg \cdot kg^{-1}$), Zn (207.77 $mg \cdot kg^{-1}$) and Cr (447.50 $mg \cdot kg^{-1}$). Both (0.01 M $Na_2S_2O_5$), ($Na_2S_2O_5$) + (0.1 HCl), and Distilled water were found to be effective on removing ($Na_2S_2O_5$) Pb, Cr and Zn respectively. ($Na_2S_2O_5$) + (0.1 HCl) Cr, Zn and Pb respectively. Sulfur Pb, Cr and Zn respectively. The removal rate of Pb, Zn, and Cr varied from 10.35% - 26%, 3.4% - 21.60% and 4.97% - 23.88% for (0.01 M $Na_2S_2O_5$) respectively. The removal rate of Pb, Zn, and Cr varied from 16.13% - 20.07%, 8.20% - 23.48%, 5.42% - 28.93% for (0.01 M $Na_2S_2O_5$ + 0.1 M HCl) respectively. The removal rate of Pb, Zn, and Cr varied from 10.20% - 25.5%, 9.55% - 25.13% and 6.04% - 25.54% for (S) respectively.

Keywords

Sodium Metabisulfite ($Na_2S_2O_5$), HCl, Sulfur and Distilled Water, Pb, Zn and Cr Soil Contaminated Sequential Washing

1. Introduction

Industrialization and modern lifestyles have led to the production and release of hazardous effluents into the environment [1] [2]. Pollution of soils with nonbiode-gradable pollutants such as heavy metals is an important environmental concern that affects both industrialized and emerging countries [3] [4] [5] [6]. It

has been estimated that about 235 million ha of arable land worldwide are polluted by heavy metals [7]. Heavy metals have a considerable toxicity for microorganisms, plants, and animals and can present high potential risks to human health [8] [9] [10]. Many chemical, physical, and biological methods have been suggested to solve the problem of heavy metal contamination, in which soil washing is one of the most effective treatments [11] [12]. Among all heavy metals, lead (Pb) has recently gained increased interest due to its potential to cause various toxicological effects in humans, such as anemia, liver and kidney damage, and cancer [13]. In soil washing techniques, where soil-bound contaminants are transferred to the liquid phase by desorption and solubilization, acid washing and chelator soil washing are two most prevalent removal methods [14]. A mixture of 0.1 M $Na_2S_2O_5$ and 0.01 M Na_2EDTA provide an economically optimum solution for Cd and Zn removal [15]. Soil washing can be practiced in two ways, batch washing and column washing. Compared to batch washing, column washing has many advantages. Firstly, the soil structure remains intact, which is not the case for batch extractions. Secondly, the process can minimize the chance of workers exposure to contaminants [16]. What's more, column washing was proved to have better metal removal effects than batch washing [17]. Soil washing is particularly frequently used in soil remediation because it: 1) completely removes the contaminants, hence ensures the rapid cleanup of a contaminated site [18], 2) meets specific criteria, 3) reduces or eliminates long-term liability, 4) may be the most cost-effective solution and may produce recyclable material or energy [19]. Since heavy metals are sparingly soluble and occur predominantly in a sorbed state, washing the soils with water alone is expected to remove too low an amount of cations in the leachates, chemical agents have to be added to the washing water [20] [21]. With chemical soil washing, soil particles are cleaned by selectively transferring the contaminants from the soil into solution [22]. The effectiveness of washing is closely related to the ability of the extracting solution to dissolve the metal contaminants in soils. However, the strong bonds between the soil and metals make the cleaning process difficult. Therefore, only extract ants capable of optimally dissolving the metals must be carefully sought during soil washing. One of the permanent solutions is soil washing with solutions containing chelating agents which, contrary to acid washing processes, permit heavy metal desorption from soil solid phase by forming strong and water soluble metal ligand complexes without deterioration of soil physico-chemical properties [23]. The object of this paper was to evaluate the removal efficiency of the heavy metal by using (0.01 M $Na_2S_2O_5$), ($Na_2S_2O_5$) + (0.1 HCl), and S+ Distilled water in treating a contaminated soil in laboratory columns and to select the optimal conditions. The study also focused on the dispose of leaching washing.

2. Materials and Methods

The clay-loam soil used in this study was collected from soil sample surface (0 -

40 cm) located in Bahira City, Egypt northern. The sampling period in (2015), Soil sampling geographic coordinate 31.197045N and 18.098445E. The sample were aired air dried and crushed pass through 2 mm sieve to elaborate the different analysis—Soil Physical Analysis: Texture was determined using sieves and Hydrometer method [24].

Soil Chemical Analysis: Salinity was measured at in the soil paste extract and pH of 1:2.5 soil suspension by EC meter and P^H [25], Sodium Absorption Ration (SAR) was calculated from Ca, Mg, and Na soluble concentrations, soil organic matter content (OM%) was determined by Walkely & Black method [25] and $CaCO_3$% was determined using the pressure calcimeter method [25]. **Table 1** shows chemical characteristics of soil surface sample extremely alkaline, p^H values > 8, Nonsaline, moderately sodic and low organic matter.

Columns washing studies were carried in lab to examine the removal efficiency of the heavy-metal by using different includes doses from (0.01 M $Na_2S_2O_5$), ($Na_2S_2O_5$) + (0.1 HCl), sulfur and Distilled water in a contaminated soil. Firstly, to evaluate the effect of (0.01 M $Na_2S_2O_5$), (0.01 $Na_2S_2O_5$) + (0.1 HCl), Sulfur (250 g) mixture with soil (2250 g) and Distilled water concentrations on the removal of Pb, Zn and Cr. Briefly, 2500 grams of contaminated soil was placed in soil columns (A, B) and 2250 grams of contaminated soil and 250 grams of sulfur of was placed in soil column (C). Distilled water was added to the column (C) samples at 980 ml ratio distilled water. The soils in the columns were initially saturated with deionized water from the bottom to avoid entrapment of air in the soil pore space. Column with 7.5 cm in diameter and 120 cm in height. A plastic mesh (D = 0.2 mm) was placed at the bottom of the column to retain the soil. Due to the long period of time required for leaching of the chelating agent solution from a high-density soil column by gravity and precipitation of some compounds, which decreased hydraulic conductivity of solution during leaching, Determination of metals: heavy metals were determined in soil sample using Atomic Absorption Spectrophotometer Shimadzu model (AA-6650). Digestion of soil sample: Using dry ashing at 430˚C - 600˚C and then diluted with 1:1 (10% HCl: Water) and then filtrate the solution.

3. Results and Discussion

Effects of $Na_2S_2O_5$ on Heavy Metals Removal:

The evolution of treatment of soil samples with a contamination rate Pb (307.31 $mg{\cdot}kg^{-1}$), Zn (207.77 $mg{\cdot}kg^{-1}$) and Cr (447.50 $mg{\cdot}kg^{-1}$) with heavy metal (Pb, Zn and Cr) Before the Additions of 0.01, $Na_2S_2O_5$ with corresponding volume

Table 1. Chemical Characteristics of soil surface sample.

Sam.	p^H	EC dS/m	Soluble Ions (meq / L)						SAR	$CaCO_3$ (%)	OM (%)
			Ca^{2+}	Mg^{2+}	Na^+	K^+	Cl^-	SO_4^{2-}			
Sample	8.49	2.49	8	7	13.8	0.4	20	3.1	5.6	13.5	1.20

of Depths in Table 2. Each for contaminated soil was equal to 980 mL per 2500 g of soil. Remediation of heavy metals contaminated soils depends, among other factors, on the time elapsing between the chelating agent in the leaching solution and the total metal content bound to the soil particles [26]. Meantime, leaching for the first, leaching (Column(A)) by $Na_2S_2O_5$ at Depth (20 cm) for Pb, Zn and Cr were 54.06 mg·kg^{-1}, 29 mg·kg^{-1}, and 60 mg·kg^{-1} respectively. at Depth (40 cm) for Pb, Zn and Cr were 31.78 mg·kg^{-1}, 27.27 mg·kg^{-1} and 20 mg·kg^{-1} respectively. at Depth (60 cm) for Pb, Zn and Cr were 42.57 mg·kg^{-1}, 44 mg·kg^{-1} and 86 mg·kg^{-1} respectively. at Depth (80 cm) for Pb, Zn and Cr were 63.78 mg·kg^{-1}, 32.53 mg·kg^{-1} and 96 mg·kg^{-1} respectively. at Depth (100 cm) for Pb, Zn and Cr were 34.97 mg·kg^{-1}, 36.63 mg·kg^{-1} and 50 mg·kg^{-1} respectively. at Depth (120 cm) for Pb, Zn and Cr were 26 mg·kg^{-1}, 33.52 mg·kg^{-1} and 90 mg·kg^{-1} respectively.

Figure 1 shows the removal efficiency Pb, Zn and Cr in soil after washing with $Na_2S_2O_5$. The removal rate of Pb, Zn and Cr varied from at Depth (20 cm) for Pb, Zn and Cr were 17.61%, 14.93% and 13.43% respectively. at Depth (40 cm) for Pb, Zn and Cr were 10.35%, 13.45% and 4.97% respectively. at Depth (60 cm) for Pb, Zn and Cr were 13.87%, 21.6% and 21.4%respectively. at Depth (80 cm)

Table 2. Heavy Metals (mg·kg^{-1}) Content in Sectioned Soil Column Leached with (0.01 M) $Na_2S_2O_5$.

Depth (cm)	Column(A)									
	Heavy Metals (mg·kg^{-1})			Removal%			Heavy Metals			
	Pb	Zn	Cr	Pb	Zn	Cr	Statistical Analysis	Pb	Zn	Cr
20	54.06	29	60	17.61	14.93	13.43	Med	38.77	33.03	73
40	31.78	27.27	20	10.35	13.45	4.97	Mean	42.19	33.82	67
60	42.57	44	86	13.87	21.6	21.4	SD	14.35	5.99	29.25
80	63.78	32.53	96	20.77	16.1	23.88	Max	63.78	44	96
100	34.97	36.63	50	11.39	18	12.44	Min	26	27.27	20
120	26.00	33.52	90	26.00	16.55	22.39				

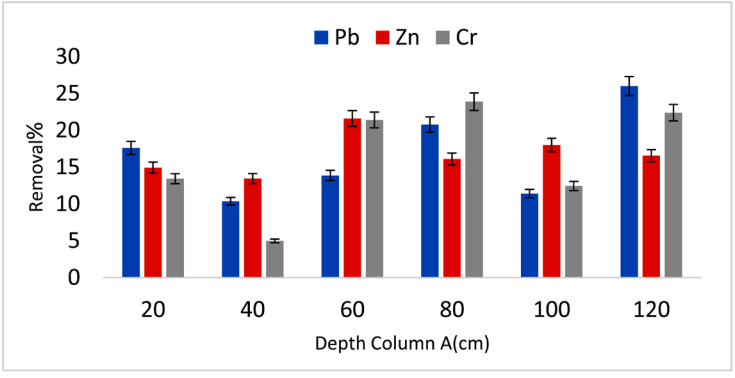

Figure 1. Effect of concentration on removal of Pb, Zn and Cr from the contaminated soil using (0.01 M $Na_2S_2O_5$).

for Pb, Zn and Cr were 20.77%, 16.1% and 23.88 %respectively. at Depth (100 cm) for Pb, Zn and Cr were 11.39%, 18% and 12.44% respectively. at Depth (120 cm) for Pb, Zn and Cr were 26.00%, 16.55% and 22.39% respectively. removal efficiencies for Pb, high at Depth (120 cm) concentration obviously affected Pb removal from contaminated soils, removal efficiencies for Cr, at Depth (80 cm), removal efficiencies for Cr, at Depth (120 cm), removal efficiencies for Zn, at Depth (60 cm), removal efficiencies for Zn, at Depth (100 cm), removal efficiencies for pb, at Depth (20 cm). removal efficiencies for Cr and Zn respectively, low at Depth (40 cm), This phenomenon could be explained by that the presence of large concentrations of ions without participation in chelation process (such as the large quantity of sodium ions present in the $Na_2S_2O_5$ solutions) can lower the stability of metal–chelant complexes [27]. The different properties of the metals and their different release mechanisms resulted in differences in their mobility during soil washing [28] [29]. In the washing process of the column soil, initially, the weakly bound metals were released from the soil, then the residual fractions, strongly bound metals such as oxides and silicates, were extracted from the retained contaminants [30] [31].

Effects of $Na_2S_2O_5$ + HCl on Heavy Metals Removal:

The evolution of treatment of soil samples with a contamination rate Pb (307.31 $mg{\cdot}kg^{-1}$), Zn (207.77 $mg{\cdot}kg^{-1}$) and Cr (447.50 $mg{\cdot}kg^{-1}$) with heavy metal (Pb, Zn and Cr) Before the Additions of $Na_2S_2O_5$+HCl with corresponding volume of Depths in Table 3 Each for contaminated soil was equal to 980 mL per 2500 g of soil. Washing of metal-contaminated soils with 0.1 M HCl, showed some promise as a remediation technique as it rapidly and effectively removes some heavy metals (e.g., Cr, Pb, Zn and Ni) from contaminated soils [32]. Remediation of heavy metals contaminated soils depends, among other factors, on the time elapsing between the chelating agent in the leaching solution and the total metal content bound to the soil particles [26]. Meantime, leaching for the first, leaching (Column (B)) by $Na_2S_2O_5$ + HCl at Depth (20 cm) for Pb, Zn and

Table 3. Heavy metals ($mg{\cdot}kg^{-1}$) Content in sectioned soil column leached with $Na_2S_2O_5$ + HCl.

Depths (cm)	Column(B)									
	Heavy Metals ($mg{\cdot}kg^{-1}$)			Removal%			Heavy Metals			
	Pb	Zn	Cr	Pb	Zn	Cr	Statistical Analysis	Pb	Zn	Cr
20	45	23.45	6.70	16.13	14.68	11.54	Med	46	26.055	9.50
40	50	13.11	3.15	17.92	8.20	5.42	Mean	46.5	26.62	9.67
60	36	23.62	16.78	12.90	14.78	28.93	SD	6.5	8.62	4.68
80	56	37.51	12.38	20.07	23.48	21.34	Max	56	37.51	16.78
100	45	28.49	9.02	16.13	17.83	15.55	Min	36	13.11	3.15
120	47	33.54	9.97	16.84	21	17.19				

Cr were 45 mg·kg^{-1}, 23.45 mg·kg^{-1} and 6.70 mg·kg^{-1} respectively. At Depth (40 cm) for Pb, Zn and Cr were 50 mg·kg^{-1}, 13.11 mg·kg^{-1} and 3.15 mg·kg^{-1} respectively. At Depth (60 cm) for Pb, Zn and Cr were 36 mg·kg^{-1}, 23.62 mg·kg^{-1} and 16.78 mg·kg^{-1} respectively. At Depth (80 cm) for Pb, Zn and Cr were 56 mg·kg^{-1}, 37.5 mg·kg^{-1} and 12.38 mg·kg^{-1} respectively. At Depth (100 cm) for Pb, Zn and Cr were 45 mg·kg^{-1}, 28.49 mg·kg^{-1} and 9.02 mg·kg^{-1} ppm respectively. At Depth (120 cm) for Pb, Zn and Cr were 47 mg·kg^{-1}, 33.54 mg·kg^{-1} and 9.97 mg·kg^{-1} respectively.

Figure 2 shows the removal efficiency Pb, Zn and Cr in soil after washing with $Na_2S_2O_5$ + HCl. The removal rate of Pb, Zn and Cr varied from at Depth (20 cm) for Pb, Zn and Cr were 16.13%, 14.68% and 11.54% respectively. at Depth (40 cm) for Pb, Zn and Cr were 17.92%, 8.20% and 5.42% respectively. at Depth (60 cm) for Pb, Zn and Cr were 12.90%, 14.78% and 28.93% respectively. at Depth (80 cm) for Pb, Zn and Cr were 20.07%, 23.48% and 21.34% respectively. at Depth (100 cm) for Pb, Zn and Cr were 16.13%, 17.83% and 15.55% respectively. at Depth (120 cm) for Pb, Zn and Cr were 16.84%, 21% and 17.19% respectively. removal efficiencies for Cr, high at Depth (60 cm) concentration obviously affected Cr removal from contaminated soils, removal efficiencies for Zn, at Depth (80 cm), removal efficiencies for Pb, at Depth (80 cm), removal efficiencies for Pb, at Depth (40 cm), removal efficiencies for Zn, at Depth (100 cm), removal efficiencies for pb, at Depth (20 and 100 cm). removal efficiencies for Zn and Cr respectively, low at Depth (40 cm). 0.1 M hydrochloric acid releases heavy metals from many soil "pools", e.g., exchangeable, organically complexed and secondary minerals (carbonates, oxides and hydroxides of iron and manganese) [33]. HCl is very effective in the removal of heavy metals and achieve high removal efficiency in the case of natural pH without neutralization to no loss the acidic property that are of high importance for the movement of contaminant sand solubility and thus easily extracted.

Effects of Sulfur 250 g (10%) + Soil 2250 g (90%). Distilled water on Heavy Metals Removal:

The use of elemental S to decrease soil pH and increase the solubility of heavy

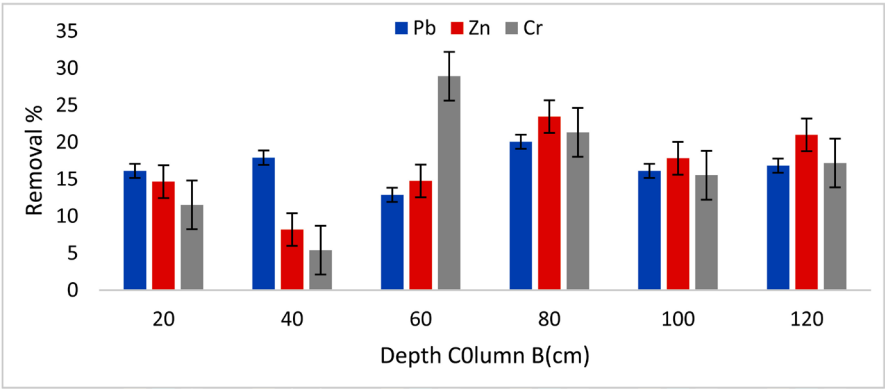

Figure 2. Effect of concentration on removal of Pb, Zn and Cr from the contaminated soil using (0.01 M $Na_2S_2O_5$ + 0.1 HCl).

metals in soils has been suggested [34] [35] [36]. However, more investigations are needed to evaluate the feasibility of this method as a tool for the enhancement of metal solubility and uptake by a metal accumulator in alkaline or neutral soils contaminated by different concentrations of heavy metals. The evolution of treatment of soil samples with a contamination rate Pb (307.31 $mg \cdot kg^{-1}$), Zn (207.77 $mg \cdot kg^{-1}$) and Cr (447.50 $mg \cdot kg^{-1}$) with heavy metal (Pb, Zn and Cr) Before the Additions of Sulfur with corresponding volume of Depths in Table 4 Each for contaminated soil was equal (Distilled water) to 980 mL per 250 g of Sulfur per of 2250 g of soil. Remediation of heavy metals contaminated soils depends, among other factors, on the time elapsing between the chelating agent in the leaching solution and the total metal content bound to the soil particles (26). Meantime, leaching for the first, leaching (Column (C)) by Sulfur at Depth (20 cm) for Pb, Zn and Cr were 7.31 $mg \cdot kg^{-1}$, 50 $mg \cdot kg^{-1}$ and 5.69 $mg \cdot kg^{-1}$ respectively. At Depth (40 cm) for Pb, Zn and Cr were 4.38 $mg \cdot kg^{-1}$, 33 $mg \cdot kg^{-1}$ and 4.14 $mg \cdot kg^{-1}$ respectively. At Depth (60cm) for Pb, Zn and Cr were 4.63 $mg \cdot kg^{-1}$, 46 $mg \cdot kg^{-1}$ and 2.22 $mg \cdot kg^{-1}$ respectively. At Depth (80 cm) for Pb, Zn and Cr were 4.31 $mg \cdot kg^{-1}$, 19 $mg \cdot kg^{-1}$ and 1.34 $mg \cdot kg^{-1}$ respectively. At Depth (100 cm) for Pb, Zn and Cr were 5.06 $mg \cdot kg^{-1}$, 21 $mg \cdot kg^{-1}$ and 4. 08 $mg \cdot kg^{-1}$ respectively. At Depth (120 cm) for Pb, Zn and Cr were 2.92 $mg \cdot kg^{-1}$, 30 $mg \cdot kg^{-1}$ and 4.80 $mg \cdot kg^{-1}$ respectively.

Figure 3 shows the removal efficiency Pb, Zn and Cr in soil after washing with Sulfur 250 g (10%) + Soil 2250 g (90%) with Distilled water. The removal rate of Pb, Zn and Cr varied from at Depth (20 cm) for Pb, Zn and Cr were 25.51%, 25.13% and 25.54% respectively. at Depth (40 cm) for Pb, Zn and Cr were 15.31%, 16.58% and 18.60% respectively. at Depth (60 cm) for Pb, Zn and Cr were 16.18%, 23.12% and 9.97% respectively. at Depth (80 cm) for Pb, Zn and Cr were 15.06%, 9.55% and 6.04 %respectively. at Depth (100 cm) for Pb, Zn and Cr were 17.71%, 10.55% and 18.32% respectively. at Depth (120 cm) for Pb, Zn and Cr were 10.23%, 15.08% and 21.54%respectively. removal efficiencies for Cr,

Table 4. Heavy metals ($mg \cdot kg^{-1}$) content in sectioned soil column leached with sulfur 250 g (10%) + Soil 2250 g (90%). Distilled water.

Depth (cm)	Column (C)									
	Heavy Metals (ppm)			Removal%			Heavy Metals			
	Pb	Zn	Cr	Pb	Zn	Cr	Statistical Analysis	Pb	Zn	Cr
20	7.31	50	5.69	25.51	25.13	25.54	Med	4.51	31.5	4.11
40	4.38	33	4.14	15.31	16.58	18.60	Mean	4.77	33.16	3.71
60	4.63	46	2.22	16.18	23.12	9.97	SD	1.44	12.7	1.63
80	4.31	19	1.34	15.06	9.55	6.04	Max	7.31	50	5.69
100	5.06	21	4.08	17.71	10.55	18.32	Min	2.92	19	1.34
120	2.92	30	4.80	10.23	15.08	21.54				

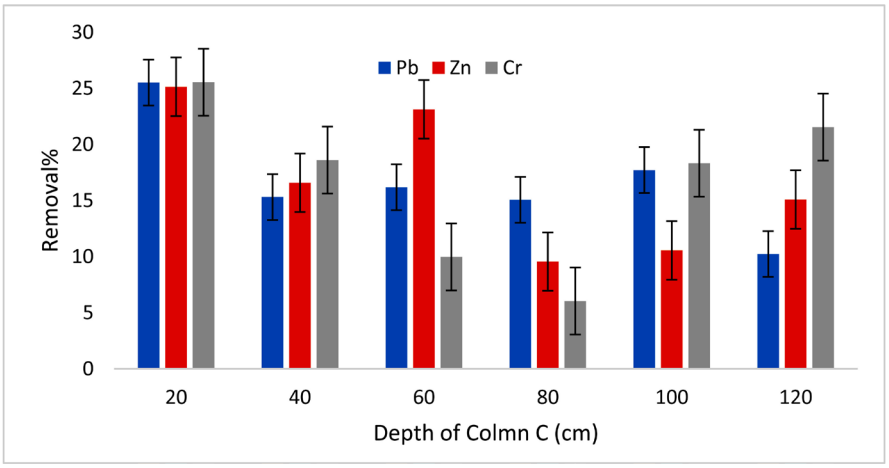

Figure 3. Effect of concentration on removal of Pb, Zn and Cr from the contaminated soil using Sulfur (10%) + Soil (90%) with Distilled water.

high at Depth (20 cm) concentration obviously affected Cr removal from contaminated soils, removal efficiencies for Pb and Zn respectively at Depth (20 cm), removal efficiencies forZn, at Depth (60 cm), removal efficiencies for Cr, at Depth (120 cm), removal efficiencies for Cr, at Depth (60 and 100 cm), removal efficiencies for pb, at Depth (20 cm). removal efficiencies for Cr respectively, low at Depths (60 and 40 cm).

4. Conclusion

The removal efficiencies of Pb, Zn and Cr in the soil by ($Na_2S_2O_5$), HCl, sulfur and distilled water at different dosages were evaluated. Both $Na_2S_2O_5$ exhibited good removal efficiencies for extractable Pb in column washing experiment, while $Na_2S_2O_5$ + HCl and concentrations of were found to be effective in extracting Cr from the contaminated soil tested in the study. But concentrations of the dosage of sulfur did not produce a proportional gain in the Pb, Zn and Cr removing from soils. Sequential extraction indicated that the elements fraction that is weakly bonded to soil particles is insignificant. That is well explained the disproportional gain with the increasing concentration of washing solution. Our study also clearly proved that sequential washing using $Na_2S_2O_5$ and $Na_2S_2O_5$ + HCl is likely to be more efficient remediation strategy for the soil than any sulfur and distilled water. This is important, since cost-effectiveness and relatively low environmental impacts are required in the on-site remediation.

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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