

Remediation of contaminated soil using soil washing-a review

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ABSTRACT

Pb, Zn, Ni, Cu, Mn and Cd are heavy metals occur naturally as trace elements in many soils. The present paper reviews the remediation of heavy metals of contaminated soil by soil washing using different agents. It was noted that the contact time, pH, concentration of extractant and agitation speed were affected the process while remediation, so accordingly select the conditions to obtain efficiency which is mainly depend upon the type of soil, contamination type, contamination period and metals present in it. EDTA is effective when compared with other chelating agents for heavy metals especially for lead but it has low biodegradation. Because of the nature of low biodegradability, EDTA can be reused further by membrane separation and electrochemical treatment, or degraded by advanced oxidation processes.

Keywords -Chelating agents, EDTA, Extractants, Heavy metals, Soil contamination, Soil washing.

I. Introduction

Nowadays, soil is becoming polluted due to some form of industrial activity, agricultural chemicals or the improper disposal of waste. The biggest concern associated with soil contamination is the harm that it can cause to human health and environment. To remediate a soil, there are a number of different methods currently employed in the process of dealing with soil contamination. Mainly, the goal of soil remediation is to restore the soil to its natural, pollution-free state. This study deals with soil washing using different extractants. The advantage of soil washing method is the high extraction efficiency and the specificity for heavy metals. To achieve treatment costs low, the soil can be reused and it should be possible to recover and reuse the chelating agent for further extraction cycles, so that soil cleanup achieved.[1]

Soil washing has the following merits. [2][3]

- Reducing the volume of decontaminant soil, so thus reducing costs;
- Physical separation allows simultaneous treatment of metal and organic pollutants;
- Metals can be recovered and reused;
- Treated soil can be redeposited on site;
- Metals adsorbed to soil particles can be treated;
- Generally removal of pollutant efficiency is between 60-90%;
- Costs are relatively low.

The main disadvantages of soil by washing are:

- For the equipment, the need of a space big enough;
- The need for wash water treatment;

- The use of washing agents increase the cost of the process and may cause problems related to reuse of treated soil;
- The technology is not suitable for soils with high clay content [2][3]

II. Sources of contamination

There are two types of contamination, namely:

2.1. *Artificial contamination* which is contamination made by artificially in order to get good homogeneity in terms of consistent heavy metals concentration and speciation, soil composition, contamination process and contamination period. This would minimize ambiguity in the extraction results arising from sample heterogeneity. [4]

2.2. *Naturally contaminated soil.* The natural contamination of heavy metals was occurred due to many activities such as Fertilizers, Pesticides, Bio solids and Manures, Wastewater, Metal Mining and Milling Processes and Industrial Wastes and Air-Borne Sources. Use of all these, can unknowingly occurs heavy metals such as Pb, Cd, Hg, Ni and Zn etc., in the soil and cause effects on the environment.

III. Soil washing using different extractants

Several types of chemical agents can be used to extract heavy metals for soil washing technology from contaminated soils. The extractants may be acids, bases, chelating agents, electrolytes, oxidizing agents and surfactants [1][5][6][7], but the most popular extractive reagents are acids and chelating agents for heavy metals decontamination.

To enhance metal extraction, chelating agents, acids, and reducing/oxidizing (redox) agents are most applicable. Chelating agents extract metals from soils by forming highly stable and soluble metal complexes that tend to dissociate from the sorption sites on the soil surfaces [8][9]. Strong acids (e.g., hydrochloric acid, phosphoric acid, sulphuric acid, and nitric acid) extract metals by dissolving discrete/surface metal precipitates and soil minerals. Reducing agents such as hydroxylamine hydrochloride (NH₂OH-HCl) and sodium bisulphite (Na₂S₂O₅) lead to dissolution of Fe/Mn oxides, thus increases the extraction of metals bound to these oxides. Oxidizing agents such as potassium permanganate (KMnO₄) and sodium hypochlorite (NaClO) facilitate chromium removal by a conversion of trivalent chromium, which is readily sorbed or precipitated, to hexavalent chromium, which is soluble [3][10].

It is recommended that an ideal chemical agent would: (i) interact very weakly with the soil matrix as compared to the target contaminants, (ii) enhance the solubility and mobility of the target contaminants, and (iii) be non-toxic and biodegradable [11]. It is, however, also recognized that it is nearly impossible to have a single chemical agent that possesses all these desirable characteristics. Despite the proven efficiency of acid extraction in full-scale applications for non-calcareous soils, strong acids result in significant dissolution of soil minerals and organic matter (up to 50%), but destructs the basic nature of soil and soil structure, also together increase the soil acidity. It causes severe damage to physical, chemical, and biological properties of soils so limits the suitability of the treated soils for being returned to the site [3][10][12][13][14]. On the other hand, chelating agents are capable of extracting metals with much less impact on soils. Therefore, soil-related research on chelating agents has increased due to the proposed use of chelating agents for soil remediation [15].

The best known chelating agents are EDTA (ethylenediaminetetraacetic acid), NTA (nitrilotriacetic acid), and DTPA (diethylenetriaminepentaacetic acid) [15].

The European authorities have conducted evaluation of EDTA and the risk assessment report indicated that EDTA has a low toxicity profile for humans, and the environmental risks are limited to some localized cases involving high emissions at concentrations above 2.2 mg/L (about 10 µM) [16]. Although the overall loss of soil weight due to EDTA-enhanced soil dissolution may be insignificant (< 13 g kg⁻¹), dissolution of amorphous Fe and Al oxides would reduce the shear strength, destabilize the soil aggregate stability, and mobilize colloids and fine particles (soil dispersion) [17]. Moreover, bioavailability, the metal mobility and fractionation

could be modified after EDTA applications and require further investigations [18][19].

In recent years, biodegradable chelating agents such as EDDS ([S,S]ethylenediaminedisuccinic acid, which is a stereoisomer of EDTA), IDSA (iminodisuccinic acid), and MGDA (methylglycinediacetic acid) have received increasing attention [15]. In particular, EDDS has been considered a promising substitute for EDTA in soil remediation technologies. EDDS can be fully degraded in wastewater treatment [20] and in soils after an initial lag phase that is necessary for the population growth or adaptation of microbes [21][22][23]. A major disadvantage in the application of EDDS, is that EDDS cannot be recovered and reused. There is an increase in the chemical cost and whether this can be offset with a possible decrease in the cost of required treatment process needs to be justified.

IV. Analysis of metal removal efficiencies:

Table 1 gives removal efficiency of heavy metals such as Pb, Ni, Cu, Cd, Zn, As. From these studies, overall EDTA gives more efficiency to all metals when compared with other agents and especially EDTA is, good in removing lead in contaminated soil.

V. Extractants

5.1 Soil washing using EDTA

Most studies investigated the effectiveness and impacts of EDTA for soil washing and other soil remediation technologies due to its low cost and high efficacy of metal extraction [8][15][16][9]. EDTA offers the best cost and performance of all chelating agents; however, its low biodegradability, and thus its high persistence in the environment, has been a recent concern.

Extraction of heavy metals from contaminated soils using EDTA has been reported by many authors. The maximum removal efficiency of heavy metals can be achieved by using Na₂EDTA as extractant, the contact time was three hours for removing lead and four hours for removing cadmium and nickel. The pH value was 4. Maximum removal percentage of lead, cadmium and nickel in batch extraction was obtained at concentrations of 0.1 M Na₂EDTA. The sequence of heavy metals removal was found to be Cd > Pb > Ni (97, 88 and 24) % [24].

Sequential extraction procedures have been widely used to quantify the distribution of metals in contaminated soil [25][26]. In this study, it is found that the removal of metals in silty sand is more suitable than silty clay by using EDTA [27].

For removal of copper and nickel, the best match of concentration and liquid/solid was 0.3 g/L and 10 mL/g using EDTA [4].

Table 1: Heavy metals removal by applying different agents

Types of metal	Chelating agents	Contamination type	Removal efficiency (%)	Reference
Cd Pb Ni Zn Cu	EDTA	Both Artificial and Natural	97-100% 45-100% 24% 54-100% 47-98%	Hatem AselGzar et al,2015 Susantandy et al,2004 S. A. Wasay et al.1998 N. Papassiopi et al,1999 R.A. Evangelista et al,1989 H.E. Allen et al,1993 Y. Jianzhen et al,1994
Cd, Pb Ni	HCL	Both Artificial and Natural	98% 94% 55%	Hatem AselGzar et al,2015 S.R. Cline et al,1993 B.E. Reed et al,1996
Cd, Pb Ni	Acetic acid	Artificial	5.2-31.1% 42.2-100% Not exceeds 1%	Hatem AsalGzar et al,2014
Cu, Ni Zn	Texapon N-40		83.2% 82.8% 86.6%	Luis G. Torres et al., 2011
Cd Zn Cu)	Tween 80		85.9% 85.4 % 81.5%	Luis G. Torres et al., 2011
Ni Zn As	Polafix CAPB		79% 83.2% 49.7%	Luis G. Torres et al., 2011
Pb	Citric acid	Artificial	50%	Masakazu Niinae et al,2008

For metal removal of As,Cd,Zn,Pb and Cu, the study of operating variables in soil washing are discussed which included the type of EDTA,retention time, solution pH,dose,temperature,agitation,ultrasound and number of extractions using free acid EDTA,ammonium salt EDTA and disodium salt[39]. In order to obtain removal efficiency more,it is important to choose best values of above parameters. Results showed that EDTA free acid is more effective than its disodium salt and ammonium salt.

Effectiveness of EDTA was examined for the removal of lead in contaminated site[38].He suggested that removal efficiency was high when pH varies from 7 to 10 and treatment time rapidly increases for first 1h and afterwards it remains constant .Ethylenediaminetetraacetic acid appeared to offer greater potentials in remediating the high permeability soil[40].

EDTA was proved good extracting solution for Pb which has maximum solubilized concentration of Pb.[41]and recovery of lead is high using EDTA[42].The concentration of EDTA increases the removal efficiency also increases but affects the soil microorganisms and plants.[43]. Sodium EDTA are effective extraction agents for the removal of lead and zinc from silty clay soil, as well as from millpond sludge.[44]

From all these,it is observed that the efficiency of metal removal is obtained by EDTA is good which

is depend upon reaction time,pH, liquid/solid ratio and chelating agent con- centration etc.By choosing an accurate of all these factors can provides more efficiency.However,EDTA has a disadvantage of low bio degradability,but it is possible to reuse again.

Therefore, risk reduction measures should be considered to ensure that no residual EDTA is allowed to remain in the treated soils or to leach into the environment after soil remediation. At the end of soil washing, the processed soils should be rinsed with clean water to remove residual EDTA, and the washing solution should be treated to remove or recover EDTA. Additions of ferric chloride, sodium phosphate, calcium hydroxide, and pH adjustment have been shown to effectively recover both metals and EDTA by precipitation and sedimentation [45]. On the other hand, metal-EDTA complexes can be separated from the washing solution by membrane separation and electrochemical treatment, or degraded by advanced oxidation processes [46][47], although these treatments are more costly than chemical precipitation

5.2 Soil washing using different agents

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 [24].The maximum removal efficiencies were 98, 94 and 55% of Cd, Pb and Ni

respectively with the best contact time was four hours for removing lead and cadmium, and five hours for nickel using concentrations 1 M HCl.

Acetic acid is effective in removing Lead and moderately effective in cadmium. Solubility of Ni in Acetic acid was very low, hence it is failed to reduce nickel in contaminated soil [36].

Compared different surfactants and also with tap water [37]. Surfactants with average removals (this is, the removal for all the metals studied) of 67.1% (Tween 80), 64.9% (Surfactopol 14104) and 61.2% (Emulgin W600.) and there were exceptional removals using Texapon N-40 (83.2%, 82.8% and 86.6% for Cu, Ni and Zn), Tween 80 (85.9, 85.4 and 81.5 for Cd, Zn and Cu), PolafixCAPB (79%, 83.2% and 49.7% for Ni, Zn and As). The worst results were obtained with POLAFIX LO with a global removal of 45%, well below of the average removal with tap water (50.2%). It is suitable for soil contaminated with organic compound.

VI. Conclusion

This review compares the different extractants for heavy metals removal in the contaminated soil. From these studies, it is concluded that EDTA is more effective on removal of heavy metals and also excellent solubilizing agents for metals including lead. However, EDTA is quite persistent in the environment due to its low biodegradability. So, EDTA can be reused again and also it does not damage the nature of soil whereas acids causes changes in the properties of soil.

References

- [1] J. J. Hong, S. M. Yang, Y. K. Choi and C. H. Lee, (1995) "Pre- cipitation of Tricarboxylic Acid Biosurfactant Derived From Spiculisporic Acid with Metal Ions in Aqueous Solution," *Colloid and Interface Science*, Vol. 173, No. 1, 1995, pp. 92-103. doi:10.1006/jcis.1301
- [2] Mann M. J., Dahlstrom D., Peterson G., et al, (1993) *Innovative Site Remediation Technology Soil Washing/Soil Flushing*, vol. 3, American Academy of Environmental Engineers
- [3] Dermont, G., Bergeron, M., Mercier, G. and Richer-Lafleche, M. (2008). "Soil washing for metal removal: A review of physical/chemical technologies and field applications." *Journal of Hazardous Materials*, 152, 1-31
- [4] Wei Jiang, Tao Tao, Zhiming Liao (2011) Removal of Heavy Metal from Contaminated Soil with Chelating Agents, *Open Journal of Soil Science*, 2011, 1, 70-76
- [5] O. Schramel, B. Michalke and A. Kettrup, (2000) "Study of the Copper Distribution in Contaminated Soils of Hop Fields by Single and Sequential Extraction Procedures," *The Science of the Total Environment*, Vol. 263, No. 1-3, pp. 11-22. doi:10.1016/S0048-9697(00)00606-9
- [6] K. R. Reddy and S. Chinthamreddy, (2000) "Comparison of Ex- tractants for Removing Heavy Metals from Contaminated Clayey Soils," *Soil and Sediment Contamination*, Vol. 9, No. 5, pp. 449-462. doi:10.1080/10588330091134347
- [7] B. Sun, F. J. Zhao, E. Lombi and S. P. McGrath, (2001) "Leach- ing of Heavy Metals from Contaminated Soils Using EDTA," *Environmental Pollution*, Vol. 113, No. 2, pp. 111-120. doi:10.1016/S0269-7491(00)00176-7
- [8] Nowack, B. (2002). "Environmental chemistry of aminopolycarboxylate chelating agents." *Environmental Science and Technology*, 36, 4009-4016
- [9] Lestan, D., Luo, C. L. and Li, X. D. (2008). "The use of chelating agents in the remediation of metal-contaminated soils: A review." *Environmental Pollution*, 153, 3-13.
- [10] Peters, R. W. (1999). "Chelant extraction of heavy metals from contaminated soils." *Journal of Hazardous Materials*, 66, 151-210
- [11] Vulava, V. M. and Seaman, J. C. (2000). "Mobilization of lead from highly weathered porous material by extracting agents." *Environmental Science and Technology*, 34, 4828-4834
- [12] Reed, B. E., Carriere, P. C. and Moore, R. (1996). "Flushing of a Pb (II) contaminated soil using HCl, EDTA, and CaCl₂." *Journal of Environmental Engineering*, 122, 48-50.
- [13] Neale, C. N., Bricka, R. M., and Chao, A. C. (1997). "Evaluating acids and chelating agents for removing heavy metals from contaminated soils." *Environmental Progress*, 16(4), 274-280
- [14] Davis, A. P. and Hotha, B. (1998). "Washing of various lead compounds from a contaminated soil column." *Journal of Environmental Engineering*, 124, 1066-1075.
- [15] Nowack, B. and VanBriesen, J. M. (eds.) (2005). *Biogeochemistry of Chelating Agents*. ACS Symposium Series Vol. 910; American Chemical Society: Washington, DC.
- [16] Nowack, B., Schulin, R. and Robinson, B. H. (2006). "A critical assessment of chelant-enhanced metal phytoextraction." *Environmental Science and Technology*, 40, 5225-5232

- [17] Tsang, D. C. W., Zhang, W. and Lo, I. M. C. (2007b). "Copper extraction effectiveness and soil dissolution issues of EDTA-flushing of artificially contaminated soils." *Chemosphere*, 68, 234-243.
- [18] Udovic, M. and Lestan, D. (2009). "Pb, Zn and Cd mobility, availability and fractionation in aged soil remediated by EDTA leaching." *Chemosphere*, 74, 1367-1373.
- [19] Zhang, W., Huang, H., Tan, F., Wang, H., and Qiu, R. (2010). "Influence of EDTA washing on the species and mobility of heavy metals residual in soils." *Journal of Hazardous Materials*, 173, 369-376.
- [20] Vandevivere, P. C., Saveyn, H., Verstraete, W., Feijtel, T. C. J. and Schowanek, D. R. (2001a). "Biodegradation of metal-[S.S]-EDDS complexes." *Environmental Science and Technology*, 35, 1765-1770
- [21] Tandy, S., Ammann, A., Schulin, R. and Nowack, B. (2006). "Biodegradation and speciation of residual SS-ethylenediaminedisuccinic acid (EDDS) in soil solution left after soil washing." *Environmental Pollution*, 40, 2753-2758
- [22] Wang, G., Koopmans, G. F., Song, J., Temminghoff, E. J. M., Luo, Y., Zhao, Q. and Japenga, J. (2007). "Mobilization of heavy metals from contaminated paddy soil by EDDS, EDTA, and elemental sulfur." *Environmental Geochemistry and Health*, 29, 221-235.
- [23] Meers, E., Tack, F. M. G. and Verloo, M. G. (2008). "Degradability of ethylenediaminedisuccinic acid (EDDS) in metal contaminated soils: Implications for its use soil remediation." *Chemosphere*, 70, 358-363.
- [24] Hatem AsalGzar,Israa Mohammed Gatea, (2015)Extraction of heavy metals from contaminated soils using EDTA and HCl.*Journal of Engineering*,Volume 21 January.
- [25] A. Tessier," P. G. C. Campbell, and M. Bisson (1979) Sequential Extraction Procedure for the Speciation of Particulate Trace Metals, *ANALYTICAL CHEMISTRY*, VOL. 51, NO. 7, JUNE
- [26] G.E.M. Hall, J.E. Vaive, R. Beer, M. Hoashi, (1996) Selective leaches revisited, with emphasis on the amorphous Fe oxyhydroxide phase extraction ', *Journal of Geochemical Exploration* 56 59-78
- [27] Krishna R.Reddy,SwapnaDanda,YelizYukselen-Aksoy and Ashraf Z.Al-Hamdan(2010) Sequestration of heavy metals in soils from two polluted industrial sites: implications for remediation,*Land Contamination & Reclamation*, 18 (1).
- [28] Susantandy, Karin Bossart , Roland Mueller , Jens Ritschel , Lukas Hauser , Rainer Schulin , and Bernd Nowack (2004) Extraction of Heavy Metals from Soils Using Biodegradable Chelating Agents,*Environ.Sci Technology* . ,38,937-944
- [29] S. A. Wasay, S. F. Barrington & S. Tokunaga (1998)Remediation of Soils Polluted by Heavy Metals using Salts of Organic Acids and Chelating Agents,*Environmental Technology* ,Vol 19,pp 369-380
- [30] N. Papassiopi, S. Tambouris and A. Kontopoulos,(1999)REMOVAL OF HEAVY METALS FROM CALCAREOUS CONTAMINATED SOILS BY EDTA LEACHING,*Water, Air, and Soil Pollution* 109: 1-15.
- [31] R.A. Evangelista, A.P. Zownir, (1989)Lead contamination removal by soil washing, *Air and Waste Management Assoc., The 82nd Annual Meeting and Exhibition, Anaheim, CA, June 25-30*, pp. 2-16.
- [32] H.E. Allen, P. Chen,(1993) Remediation of metal contaminated soil by EDTA, *Environ. Prog.* 12 4 284-293.
- [33] Y. Jianzhen, D. Klarup,(1994) Extraction kinetics of copper, zinc, iron and manganese from contaminated sediment using disodium ethylenediaminetetraacetate, *Water, Air, Soil Pollut.* 75 1994 205-225.
- [34] S.R. Cline, B.E. Reed, M.R. Matsumoto,(1993) Efficiencies of soil washing solutions for the remediation of lead contaminated soils, *Haz. Ind. Wastes. Proc. Mid. Atl. Ind. Waste Conf. College Park, MD*, pp. 169-177.
- [35] B.E. Reed, P.C. Carriere, R. Moore,(1996) Flushing of a Pb II contaminated soil using HCl, EDTA, and CaCl₂ , *J. Environ. Eng., ASCE* 122 1 1996 48-50.
- [36] Hatem AsalGzar et al.,(2014)Extraction of Lead, Cadmium and Nickel from Contaminated Soil Using Acetic Acid, *Open Journal of Soil Science*, 2014, 4, 207-214
- [37] Luis G. Torres , Rosario B. Lopez , Margarita Beltran,(2011) Removal of As, Cd, Cu, Ni, Pb, and Zn from a highly contaminated industrial soil using surfactant enhanced soil washing, *Physics and Chemistry of the Earth*
- [38] Masakazu Niinae, KoudaiNishigaki* and Kenji Aoki,(2008)Removal of Lead from Contaminated Soils with Chelating

- Agents, Materials Transactions, Vol. 49, No. 10 pp. 2377 to 2382
- [39] Zeli Zou.(2008)The study of operating variables in soil washing with EDTA,Environmental pollution 157 (2009) 229–236,2008.
- [40] Wuana, R. A, Okieimen, F. E, Imborvungu,(2010) J. A, Removal of heavy metals from a contaminated soil using organic chelating acids, Int. J. Environ. Sci. Tech., 7 (3), 485-496, summer 2010
- [41] Iram Shazia and Ul-Hassan Mahmood (2014), Effect of Chelating agents on Heavy Metal Extraction from Contaminated Soils, Research Journal of Chemical Sciences Vol. 4(9), 70-87, September
- [42] H. A. Elliott and G. A. Brown, comparative evaluation of NTA and EDTA for Extractive decontamination of Pb-polluted soils,(1989)Water, Air, and Soil Pollution 45: 361-369.
- [43] Bibhabasu Mohanty and Amit. B. Mahindrakar,(2011)Removal of Heavy Metal by Screening Followed by Soil Washing from Contaminated Soil,- International Journal of Technology And Engineering System(IJTES)-Vol.2.No.3.
- [44] Riyad J. Abumaizar Edward H. Smith,(1999)Heavy metal contaminants removal by soil washing,Journal of Hazardous Materials B70 1999 71–86
- [45] Lo, I. M. C. and Zhang, W. (2005). “Study on the optimal conditions for EDTA recovery from soil washing effluents.” Journal of Environmental Engineering, 131, 1507-1513.
- [46] Finzgar, N. and Lestan, D. (2008). “The two-phase leaching of Pb, Zn and Cd contaminated soil using EDTA and electrochemical treatment of the washing solution.” Chemosphere, 73, 1484–1491.
- [47] Pocięcha, M., Lestan, D. (2009). “EDTA leaching of Cu contaminated soil using electrochemical treatment of the washing solution.” Journal of Hazardous Materials, 165, 533–539.