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Reclamation of Lead and Cadmium Contaminated Soil Using Soluble Organic Matter

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ABSTRACT

The present study reports the feasibility of reclamation of a lead and cadmium contaminated soil using the solution containing organic carbon resulting from the decomposition of organic matter, an environmentally-friendly and cost-effective leachate. For conducting this experiment, three soil samples were used. Column leaching experiments were performed to test the efficiency of organic solution containing two rates of organic carbon (0.25 g/L and 0.50 g/L) for the reclamation of the lead and cadmium contaminated soil in natural conditions. Results showed that utilization of the solution containing organic carbon resulting from the decomposition of organic matter was effective for removal of lead from the contaminated soil, attaining 30.87%-74.97% of Pb in an alkaline condition. On the other hand, it was found that this concentration of carbon was not much effective for Cd removal from contaminated soil, attaining only 1.18%-24.14% of Cd. Clay content, pH and organic matter content of soil might be responsible for such differences in removal efficiency among samples. Such a reclamation procedure indicated that solution containing organic carbon released during decomposition of fresh organic matter is a promising agent for remediation of lead contaminated soils. However, further research is needed before the method can be practically used for *in situ* reclamation of heavy metal contaminated sites.

Keywords

Soil contamination,
Heavy metals, Organic
carbon, Leachate,
Reclamation

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Introduction

Contamination of soils with toxic metals has become a major environmental concern in many parts of the world due to rapid industrialization, increased urbanization, modern agricultural practices and inappropriate waste disposal methods (Flathman and Lanza, 1998). In the past, soil contamination was not considered as important as air and water pollution, because soil contamination was more difficult to be

controlled and governed than air and water pollution. It is highly desirable to apply suitable remedial approaches to polluted soil, which can reduce the risk of metal contamination. The excavation and disposal of soil is no longer considered to be a permanent solution. The demand for soil a treatment technique is consequently growing and the development of new low-cost, efficient and environmental friendly remediation technologies has generally become one of the key research activities in environmental

science and technology. In selecting the most appropriate soil remediation methods for a particular polluted site, it is of paramount importance to consider the characteristics of the soil and the contaminants (Domen *et al.*, 2008).

Remediation methods of contaminated soils with heavy metals can be roughly classified into physical or chemical, and phytoremediation (Zhou and Song, 2004). Remediation mechanisms basically consist of two fundamental principles. The first is to completely remove contaminations from polluted sites and the second is to transform these pollutants to harmless forms by using one or more engineering technologies, which mainly include excavation, separation, extraction, electrokinesis, washing, oxidation, reduction, phytoextraction, phytovolatilization, or solidification, vitrification etc. (Aboulroos *et al.*, 2006), (Chen *et al.*, 2006). Among these developed remediation technologies, soil washing process gives high removal efficiency for remediating sites contaminated with heavy metals using suitable chelating agents, surfactant, acids, alkalis and complexing agents because it can be applied to large contaminated areas due to its rapid kinetics, operational easiness, and economical efficiency. 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 (Davies and Singh, 1995; Dikinya and Areola, 2010).

Several classes of chemicals used for soil washing include surfactants, cosolvents, cyclodextrins, chelating agents and organic acids (USEPA, 1990; Wood *et al.*, 1990; Chu and Chan, 2003; Gao *et al.*, 2003; Maturi and Reddy, 2008; Zhang *et al.*, 2009; Zvinowanda *et al.*, 2009). However, solution released from

decomposed organic matter which contains organic compounds possess the same characteristics as any chelating agents to decontaminate the metal contaminated soils by making complex with the metals like any chelating agent may result more higher efficiency to decontaminate the soil than any other chelating agents (Liu and Lin, 2013). Based on the concept of ion exchange, the solution released from decomposed organic matter is expected to suppress the desorption of original nutrients in the soil during washing owing to its high nutrient content. The constituents of the solution may act as chelating agents. It can also be used to enrich the soil. These methods can be used to prevent further contamination of soil when other treatment options are not physically or economically feasible for a site (Peters, 1999), (Xu and Zhao, 2005).

The objectives of the research were to highlight on the reclamation of heavy metal contaminated soils

To determine the favorable treatment for remediation by organic solution.

To find out an environmental friendly and simple method for remediation of particular metal contaminated soils in Bangladesh.

Materials and Methods

Sample collection and preparation

Composite soil samples were collected from the agricultural fields along the Karnatali and Dhaleshwari river which are exposed to the different degrees of environmental pollution from nearly industrial effluents and wastes. In Karnatali sampling area, two soil samples were collected from two different sampling points and one soil sample was collected from Dhaleswari sampling area a sampling and both areas were irrigated with river water. In every

sampling point, samples were collected from surface (depth up to 15 cm) using an auger as outlined by FAO (1980) and Jackson (1962). Each of the collected soil samples were air dried and passed through a 2 mm stainless steel sieve for leaching experiment. A portion of the soil sample (2 mm sieved) was further ground and passed through a 0.5 mm sieve for chemical analysis. Cation exchange capacity was done with 2mm sieved soil.

Mixed vegetables were collected from uncontaminated agricultural fields and left it for some days for decomposition. After 15 days of decomposition process, liquid was released from the decomposing organic matter (e.g. vegetables) was collect and preserved for soil washing experiments.

Laboratory analysis of the samples

Before conducting the washing experiment and incubation study, various physical and chemical properties of soil samples and organic matter were determined. Among physical properties, particle size analysis of the soil was carried out by hydrometer method as described by Gee and Bauder (1986). The textural classes were determined by Marshall's triangular co-ordinates as derived by the United States Department of Agriculture (USDA, 1951).

Soil pH was measured (field condition) electrochemically by using Jenway (Model 40) glass electrode pH meter. The cation exchange capacity (CEC) of soil was determined by ammonium acetate extraction method as described by Schollenberger and Simon (1945). The electrical conductivity of the soil was measured in the saturation extract of the soil with the help of an EC meter.

The organic carbon of the soil samples was determined by Walkley and Black's (1934) wet - oxidation method as outlined by Jackson

(1973). Soil organic matter was calculated by multiplying the percent value of organic carbon with the conversional factor of 1.724 and total organic carbon of the liquid sample that released from the decomposition of organic matter was determined by Tyurin's method (Tyurin, 1931).

Total nitrogen was determined by micro Kjeldahl's method as described by Jackson (1973). The total phosphorus content of the samples was determined by vanadomolybdophosphoric yellow color method using a spectrophotometer (Jackson, 1973) from HNO₃-HClO₄ digest as described by Piper (1947).

The total potassium content of the samples was measured by flame analyzer (Jenway, 1973).

Total cadmium (Cd), lead (Pb), Chromium (Cr), nickel (Ni) and zinc (Zn) content of the soil and organic matter were determined directly by using Atomic Absorption Spectrophotometer (Varian AA240) from nitric acid (HNO₃) digest.

Soil washing experiment

A column leaching experiment was set up with metal spiked soils and the liquid that released from the decomposition of organic matter.

Incubation of soil sample

The soils were artificially spiked with 100 µg/ml lead nitrate (PbNO₃) and cadmium nitrate (CdNO₃) solution maintaining 30% moisture. The soil and metal salt solution was allowed to react for three days in moist condition (30% moist). A control treatment (without adding metal salt solution) was also included. All the treatments were replicated twice.

Washing the soil column by leachate

The soil washing experiment was conducted in a column leaching process using two concentrations (0.25 g/L and 0.50 g/L) carbon of the liquid that released during the decomposition of organic matter. For this experiment, a leaching tube of 25 cm was used. 10gm of soil sample was kept in the leaching tube. Then successive leaching was conducted by maintaining organic carbon concentration of 0.25 g/L and 0.50 g/L. Each time 50 ml of liquid solution was used to leach the soil. A total of three times successive leaching was done. When the leaching of liquid through the soil was completed, the leachate was collected and digested to determine the metal concentration in the leachate by using Atomic Absorption Spectrophotometer (Varian AA240) following nitric acid (HNO₃) digestion. The removal efficiency of heavy metals from each soil sample was then calculated based on its initial concentration of metal in the soil prior to leaching and its final concentration of the same metal in the leachate after column leaching experiment. Experiments were conducted in duplicate and pH of the soil sample before and after leaching was also determined. However, no cadmium was found in all the three sample as background concentration. Therefore, there was no control experiment for cadmium.

Results and Discussion

Characteristics of soluble organic matter and collected soil

Organic carbon content of the organic solution was 2.82 g/L. Lead and cadmium concentration of the solution was found below detectable limit and the pH was 8.26 and 8.87 for 0.25 g/L and 0.50 g/L organic carbon containing solution respectively, which indicated that the solutions were slightly to

strongly alkaline and total nitrogen content of the solution was 0.47% and various physical and chemical properties of soil samples were presented in Table 1.

Removal efficiency for lead and cadmium

After three successive leaching of soil samples with the organic solution (0.25 g/L and 0.50 g/L organic carbon) only about 67.40%, 30.87% and 38.92% of Pb was removed from the soil 1, soil 2 and soil 3, respectively (Figure 1) and only 24.14%, 19.58% and 8.47% of Cd was removed from the soil 1, soil 2 and soil 3, respectively (Figure 3) through the application of 0.50 g/L organic carbon containing solution. Whereas application of 0.25 g/L organic carbon containing solution resulted only 31.10%, 74.97% and 60.31% of Pb was removed from the soil 1, soil 2 and soil 3, respectively (Figure 1). But application of 0.25 g/L organic carbon containing solution in three successive leaching experiments resulted only 1.18%, 1.46% and 3% of Cd removed from the soil 1, soil 2 and soil 3, respectively (Figure 3).

In case of control experiment for Pb, only about 50.65%, 44.44% and 20.57% of Pb was removed from soil 1, soil 2 and soil 3, respectively (Figure 2) through the application of 0.50 g/L organic carbon containing solution and applying 0.50 g/L organic carbon containing solution resulted 60.47%, 42.17%, and 49.23% of Pb was removed from the soil 1, soil 2 and soil 3 respectively (Figure 2).

In this investigation, the soil samples were slightly alkaline to strongly alkaline and the pH of the soil ranged from 7.22 to 8.13 (Table 1) and the pH of the organic solution (0.25 g/L and 0.50 g/L organic carbon) was 8.26 and 8.87, respectively. During leaching experiment, the pH of the soil was also alkaline and the leaching experiment was done in an alkaline condition in order to maintain

the natural condition of the soil. Therefore, pH was one of the main factors responsible for such removal efficiency in this investigation.

The chemical behavior of Pb in soil depends very much on the organic matter, clay content and pH. As it is true for all cationic metals, adsorption increased with pH. The retention of the metals did not significantly increase until the pH was greater than 7. At high pH, mobility of soil decreases. Thus, metal ability to leach through the soil decreases.

Similarly cadmium may be absorbed by clay minerals, carbonates or hydrous oxides of iron and manganese or may be precipitated as cadmium carbonate, hydroxide, and phosphate.

Evidence suggests that adsorption mechanisms may be the primary source of Cd removal from soils (Dudley *et al.*, 1991). The chemistry of Cd in the soil environment is greatly controlled by pH. Under acidic conditions Cd solubility increases and very little adsorption of Cd by soil colloids, hydrous oxides, and organic matter takes place. At pH values greater than 6, Cadmium is absorbed by the soil solid phase or is precipitated, and the solution concentrations of

cadmium are greatly reduced (McLean and Bledsoe, 1992).

Another important factor in case of cadmium is that Organic matter is important for the retention of metals by soil solids, thus decreasing mobility and bioavailability. Higher solubility of heavy metals in soil solution at alkaline pH was attributed to enhanced formation of organic matter metal complexes. Overcash and Pal (1979) reported that the order of metal-organic complex stabilities was Hg > Cu > Ni > Pb > Co > Zn > Cd which indicates that Cd is in least of this stability series and Pb will form complex more readily as compare to Cd. However, many authors have found that high organic matter content or addition of organic matter by organic wastes decreased the Cd concentration in solution.

Metals that readily form stable complexes with soluble organic matter are likely to be mobile in soils. Lead is strongly adsorbed on humic matter at pH 4 and above (Bunzl *et al.*, 1976) (Calmano *et al.*, 1993) and form dissolved Pb-organic complexes. In the pH range 7 to 10, application of organic carbon containing solution showed effective result for Pb removal (Niinae *et al.*, 2008).

Fig.1 Lead removal efficiency of organic solution (Spiked soils)

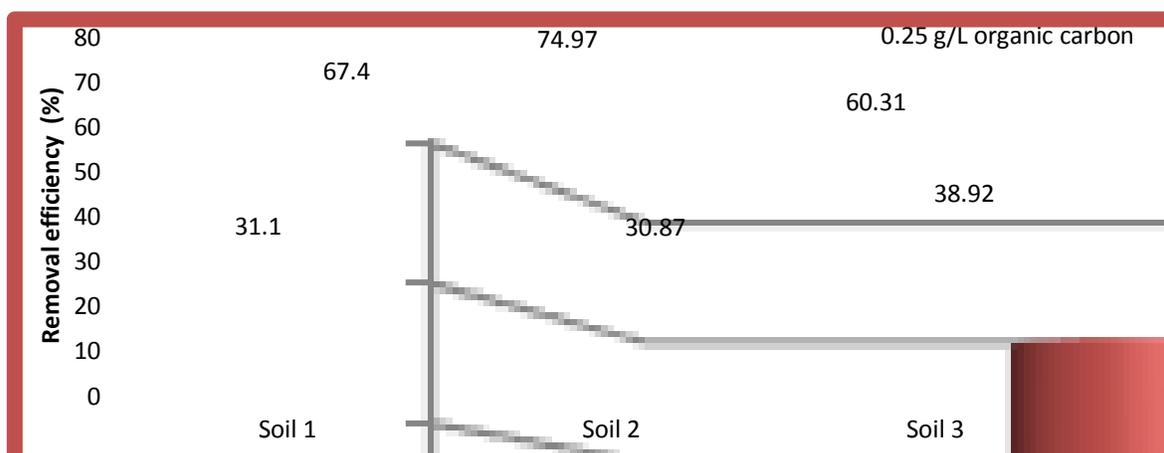


Fig.2 Lead removal efficiency of organic solution (Control)

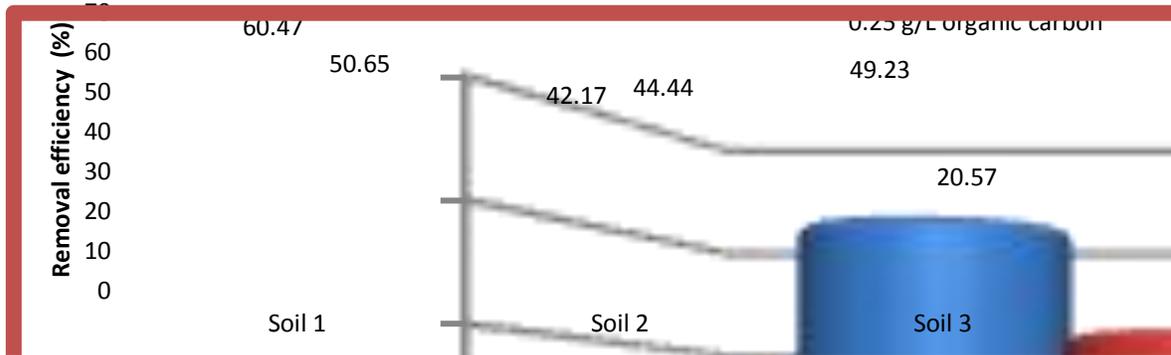


Fig.3 Cadmium removal efficiency of organic solution (Control)

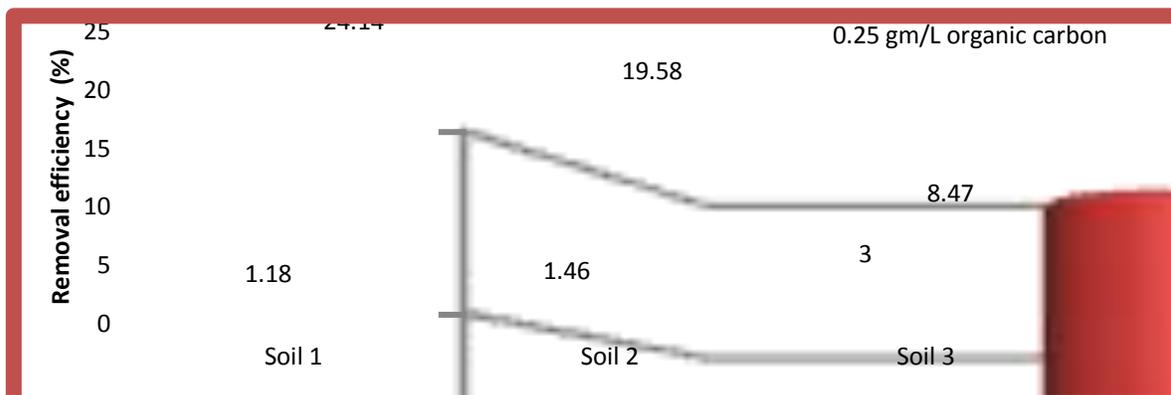


Table.1 Different properties of collected Soil

Properties	Soil 1	Soil 2	Soil 3
Sand (%)	3.17	7.69	1.23
Silt (%)	70.69	58.35	57.11
Clay (%)	26	33	42
Texture	Silt loam	Silty clay loam	Silty clay
pH	7.13	7.42	8.13
EC(ds/m)	0.38	0.19	0.40
Organic carbon (%)	0.53	0.86	0.64
Organic matter (%)	0.92	1.50	1.11
Total nitrogen (%)	0.10	0.15	0.13
Total P (%)	0.70	0.50	0.58
Total K (%)	0.23	0.23	0.28
CEC (meq/100 gm soil)	11.3	19.1	23.7
Total Pb (µg/g)	32.25	43.75	43.5
Total Cd (µg/g)	BDL	BDL	BDL
Total Zn (µg/g)	64.4	68.70	41.7
Total Cr (µg/g)	18.8	12.3	36.2
Total Ni (µg/g)	29	27	41.7

The application of 0.25 g/L organic carbon containing solution was removed highest percentage of Pb from spiked soil 2 (74.97%) and control soil 1 (60.47%) respectively. Organic matter content (1.50%) and pH (7.42) favors formation of organic complexes increases mobility of the Pb might be possible reason behind such higher removal (74.97%) of Pb from spiked soil 2.

High clay content (42%) as well as higher percentage of organic matter (1.11%) as compare to other soils might be reason for higher removal Cd from spiked soil 3 (3%) by the application of 0.25 g/L organic carbon containing solution.

In the absence of appreciable organic matter, Pb is strongly adsorbed on clay minerals (Hildebrand, 1974a) and Fe oxides (Hildebrand *et al.*, 1974b) (Kinniburgh *et al.*, 1976). Clay (26%) and organic matter (0.92%) content of soil 1 was lowest among three soils which might be the reason for such removal of highest percentage of Pb from spiked soil 1 (67.4%) and control soil 1 (50.65%) respectively by the application of 0.50 g/L organic carbon containing solution.

On the other hand, application of 0.50 g/L organic carbon containing solution resulted highest removal of Cd (24.14%) from spiked soil 1 because the pH, organic matter and clay content of soil 1 is lowest as compare to others soils may favors formation of metal soluble organic ligand complexes which increases mobility of the Cd.

Released solution from the decomposition of organic matter was used to leach the metal contaminated soil through column instead of using any commercial chemical like EDTA, citric acid or other weak acid. Application of 0.50 g/L organic carbon containing solution showed highest Pb removal efficiency from soil 1(67.40%) and highest Cd removal

efficiency from soil 1 (24.14%). On the other hand, highest Pb removal efficiency (74.97%) from soil 2 and highest Cd removal efficiency (3%) was found for soil 3 through applying 0.25 g/L organic carbon containing solution.

Application of 0.25 g/L organic carbon containing solution had higher Pb removal efficiency but it did not produce similar result for Cd removal. In case of Cd, a small amount of Cd was removed from the soil through this experiment.

Application of 0.50 g/L organic carbon solution had higher Pb removal efficiency compared to Cd removal efficiency. In general, this experiment was relatively ineffective for Cd removal because the all three soil samples were slightly alkaline and at this condition mobility of soil decreases.

Besides that concentration of organic solution, clay content and organic matter content might be the reason for these kinds of difference in removal efficiency among different soils.

On the basis of this experiment, it may therefore be recommended that the application of 0.25 g/L organic carbon containing solution was more effective for the reclamation of Pb-contaminated soil compare to the application of 0.50 g/L organic carbon containing solution.

This experiment did not show effective results in case of Cd removal in alkaline condition but application of 0.50 g/L organic carbon containing solution showed better result than 0.25 g/L organic carbon containing solution.

The results also indicates that successive application of lower concentration and higher concentration of organic carbon in leaching experiment may be effective for the removal of any cationic metals in soils.

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