



Design of Soil Remediation Techniques from Column Leaching Test Results

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The soil remediation at a contaminated site requires knowledge of contaminant transport parameters and processes. This paper presents the determination of transport parameters from column leaching tests in context with two soil remediation techniques i.e., soil washing and immobilization. To evaluate the soil washing technique, the column leaching tests on the polluted soil were conducted with diluted acid solutions of hydrochloric acid, ethylene diamine tetraacetic acid and ferric chloride to evaluate the leaching efficiencies of the selected leaching solutions. It was observed that the efficiency of diluted ferric chloride solution was higher as it removed the higher percentage of metals from the soil. From these test results, the contaminant transport parameters i.e., retardation factor and dispersion coefficient were determined which are useful to calculate the volume of leaching solution that will be required for soil washing at a site. As part of immobilization study on this soil, the soil was mixed with the selected amendments (lime, sodium hydroxide and cement) to increase the pH of the soil to 10 and the retardation factors were estimated through batch leaching test results. The retardation factors of different metals obtained with lime addition were found higher than the other amendments. To analyze the long-term stability of the amended mixtures, the leaching tests were conducted on amended soil samples and the immobilization efficiencies were estimated. It was found that the immobilization efficiencies were higher with lime addition and also concluded that the immobilization efficiencies are directly related to retardation factors.

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1. INTRODUCTION

The pollution of soil may take place due to various reasons such as unregulated disposal of wastes, mining and industrial activities, modern agricultural practices, construction activities etc. [1–3]. The heavy metal pollution of soil may create health problems for living beings as they can enter into the living beings through several ways [4]. The pollution of groundwater due to migration of heavy metals from the contaminated soil is a serious problem as the ground water will be used for various purposes including spa treatments (medicinal thermal water) [5]. For the developing or residential areas a soil remediation is very much required and before planning the remediation, it is important to conduct a survey similar to work conducted by Ezirim and Okpoechi [6].

The various soil remediation techniques in practice are immobilization, chemical treatment, electrokinetic processes, biochemical processes, soil washing, incineration techniques and thermal desorption technique [7–9]. The two important and widely used soil remediation techniques are soil washing and immobilization. Diluted acids such as hydrochloric acid (HCl) and ethylenediamine tetra acetic acid (EDTA) can be used for soil washing treatment [10–14]. The soil washing technique is effective for granular soils as the texture of the soil is more permeable. For less permeable soils such as clay, the soil washing is not very effective as the washing technique requires sufficient void spaces to wash out the pollutants from the soil. In such cases, immobilization is an alternate remediation technique where the migration of pollutants is restricted by converting the metals in soil to their stable hydroxide

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form. The soil washing require treatment of wastewater after washing the contaminants from the soil and the extracted metals need to be disposed safely [15, 16].

The immobilization technique avoids excavation of the contaminated soil, treatment of wastewater, disposal of the contaminated fluid and provides relatively cost-effective solution to treat the contaminated soil [17]. The metal hydroxides formed after immobilization will not migrate because of their low soluble form and reduces the probability of contamination of nearby water bodies [18]. Various organic and inorganic additives can be used to restrict the mobility of contaminants in the soil using immobilization technique [19–25]. The additives such as clay, calcium hydroxide, cement, zeolites, hydroxyapatite, phosphates, organic compost, microbes, activated carbon, fly ash and lime are widely used materials to immobilize the metals in soil and sludge [26–33]. Long-term stability of the amended soils also depends on the solubility of metals in their converted form [34–36]. The selection of materials or chemicals required for these soil remediation techniques depends on the transport parameters of the contaminants in the soil to be treated. In the present study, it is proposed to demonstrate the determination of contaminant transport parameters through batch tests and column leaching tests results. The one-dimensional advection-dispersion transport equation describes the rates of migration which is given in Equation (1) [37].

$$R \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2} - v_s \frac{\partial C}{\partial z} \quad (1)$$

where,

R = retardation factor = $1 + \rho K/n$

ρ = dry density (g/cm^3)

K = distribution coefficient (cm^3/g)

n = porosity

D = dispersion coefficient (cm^2/s)

v_s = seepage velocity (cm/s)

As part of the investigation, a contaminated soil was studied to know the suitability of the soil washing and immobilization techniques. The column leaching tests were carried out with 0.1 normal chemical solutions of HCl, EDTA, and Ferric Chloride (FeCl_3) on the contaminated soil and found that FeCl_3 is effective in the removal of metals. The transport rates were anticipated via preparing the elution curves using trial values substituted in theoretical equation and matching them with elution curves of experimental values. The transport rates of contaminants during column leaching tests were modeled using the MATLAB programming for the analytical solution which is based on the Shackelford and Glade's [38] leaching mass ratio (LMR) approach. Since the column leaching tests represent the leaching process in the field during soil washing, the transport rates obtained by this method are useful in the field to estimate the volume of flushing solution needed to wash out the various contaminants. The immobilization studies were

also conducted by mixing the soil with additives such as lime, sodium hydroxide and cement which increases the pH of soil and immobilize the metals in it. The amended samples were tested for their leachability to evaluate the efficiencies of additives and found that the leaching of metals from lime amended sample were less. The batch tests were performed on amended soil samples to estimate the retardation factors and found that the retardation factor of lime amended sample was higher. The values of retardation factors were compared with the efficiencies of amendments obtained from leaching tests and found that the retardation factors are useful to identify the best additive.

2. MATERIALS AND METHODS

The research methodology of the current research work is illustrated in the flow chart (Figure 1).

2.1. Characteristics of Contaminated Soil The soil samples were collected from a dumping yard located at the outskirts of Bangalore, India. The composition of the soil was estimated to be of 34% sand, 12% silt and 54% clay. The soil is classified as low plasticity clay (CL) as per Indian Standard classification system. The plasticity and compaction characteristics of the contaminated soil are as given in Table 1. The quantities of metal ions presented in the contaminated soil were estimated by following the methods specified in USEPA [39] and the estimated quantities are given in Table 2. The maximum permissible limits of heavy metals in soil specified by World Health Organization (WHO) [40] are also presented in Table 2.

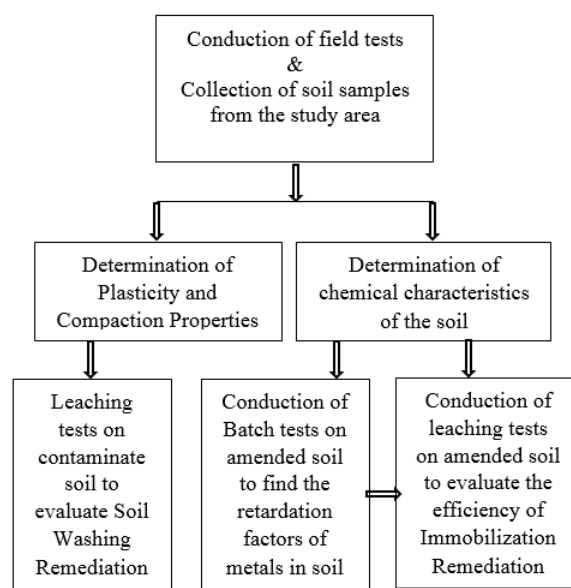


Figure 1. Flowchart of the research methodology

TABLE 1. Plasticity and compaction characteristics of contaminated soil

Specific gravity (G)	Maximum dry density (g/cc)	Optimum water content (%)	Liquid limit (%)	Plastic limit (%)	Shrinkage limit (%)	Plasticity index (Ip) (%)
2.69	1.62	24.6	56.2	24.8	13.6	31.4

TABLE 2. Heavy metals in contaminated soil

Metals in soil	Cu	Zn	Fe	Cr	Cd	Ni	Pb
Quantity of metals in soil (mg/kg)	112.8	148.6	198.7	18.9	1.2	44.6	4.8
Permissible limits of heavy metals in soil (mg/kg)	36	50	-	100	0.8	35	85

2. 2. Column Leaching Tests on Contaminated Soil

The in-situ soil washing program was simulated by performing the column leaching tests with various leaching solutions. The diluted acid solution was permitted to flow through the soil column and the effluent concentrations were measured periodically. The effluent volume (V) and the mass of contaminant leached (Δm) were found periodically. For each time interval, the leached pore volumes of flow ($T^1 = V/V_v$) were calculated. The LMR which is the ratio of leached mass of contaminant (Δm) from the soil to initial mass of contaminant (M_0) was computed periodically and the Cumulative LMR (LMR_m) was calculated with respect to time. These values were plotted with respect to their respective leached pore volumes (T^1). The plots, thus prepared are called experimental elution curves which are important to analyze the efficiency of leaching solution.

2. 3. Estimation of Transport Parameters from Column Leaching Tests

The analytical solution developed by Shackelford and Glade [38] is as given below (Equation (2)).

$$LMR_m = \frac{\sum \Delta m}{M_0} = \frac{T^1}{R} - \frac{1}{2} \left\{ \left(\frac{T^1}{R} - 1 \right) \operatorname{erfc} \left(\frac{R - T^1}{2 \sqrt{\frac{T^1 R}{P_L}}} \right) + \left(\frac{T^1}{R} + 1 \right) \exp(P_L) \operatorname{erfc} \left(\frac{R - T^1}{2 \sqrt{\frac{T^1 R}{P_L}}} \right) \right\} \quad (2)$$

LMR_m = cumulative leaching mass ratio = $\sum (\Delta m / M_0)$
 P_L = column Peclet number = $v_s L / D$;

To plot the theoretical elution curves, experimental values of T^1 and P_L along with trial values of transport parameters (D and R) were taken as input in the MATLAB program which was generated out of the above equation (Equation (2)). The theoretical elution curve was then coordinated with the experimental one to estimate the transport parameters.

2. 4. Batch Tests on Amended Soil Samples

The Batch tests were conducted to find the distribution

coefficients (K) as per ASTM specifications [41]. The distribution coefficient was then calculated from Equation (3):

$$K = \frac{(C_o - C_t)V}{M \cdot C_t} \quad (3)$$

where,

C_o and C_t respectively are initial and final concentrations of the pollutants. M and V are mass of soil taken (g) and the volume of solution used (ml) respectively. The value of K will be zero for non-reactive solutes reducing $R=1$.

2. 5. Amended Soil Samples and Leaching Tests

The contaminated soil sample of about 110 grams was taken in a container and the additive / amendment was added to the soil in such a way that the pH value of the mixture achieved the desired value. The samples were prepared with 3 inorganic additives, i.e., lime, cement and NaOH. Since the immobilization efficiencies increases with increasing pH value [27–31] and the efficiencies were observed to be higher corresponding to a pH value of 10, each selected additive was added to adjust the pH of the mixture to maintain a pH value of 10. To evaluate the long-term stability of the soil mixtures, leaching tests were conducted by passing water through the soil placed in the containers and the effluent was collected in a container. The concentrations of heavy metals in effluents were found by Atomic Absorption Spectroscopy (AAS) to know the amounts of various contaminants leached out after solidification and to assess the capabilities of these solidifying agents. From the effluent concentrations estimated, the cumulative percentage leached and the immobilization efficiency were estimated for each metal ion (Table 3).

3. RESULTS AND DISCUSSION

3. 1. Column Leaching Tests and Analytical Method

The removal efficiencies of leaching solutions studied with respect to each metal ion (copper, zinc, iron, nickel, cadmium, lead and chromium) in the sludge were

analyzed. From the column leaching tests it was observed that with diluted HCl, EDTA and FeCl₃, the removal efficiencies of metals were around 30-50%, 50-70% and 70-80%, respectively. The removal efficiencies of FeCl₃ were observed high among the three solutions. The efficiencies were in the sequence of FeCl₃ > EDTA > HCl which is similar to data reported in literature [42, 43]. The removal efficiencies were observed slightly lower than the reported data [43, 44]. This may be due to the presence of more clay fraction in the soil studied. Out of the metals studied, the removal efficiencies were in the following order which are similar to the studies conducted by Sumalatha et al. [45]. It can be observed that the removal efficiency of cadmium is the highest and that of chromium is the lowest. Cd > Pb > Zn > Cu > Fe > Ni > Cr

The elution curves obtained by washing the soil with diluted acids are shown in Figures 2 to 4. The transport parameters were found with 0.1M FeCl₃ since the higher efficiencies were achieved with this solution. The elution curves (theoretical) plotted for copper and zinc are shown in Figures 5 and 6 along with their experimental values. Similar plots were prepared for other metals and their transport parameters are reported in Table 3. The parameters thus estimated can be substituted in the MATLAB program along with the field parameters to estimate the leached pore volumes of flow required in the field and thus the volume of leaching solution needed at the site to implement the in-situ soil washing program.

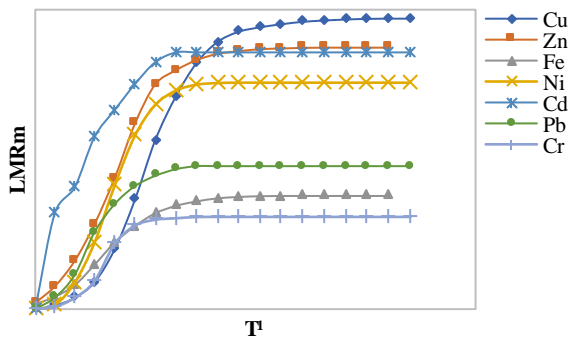


Figure 2. Elution curves (experimental) with diluted HCl

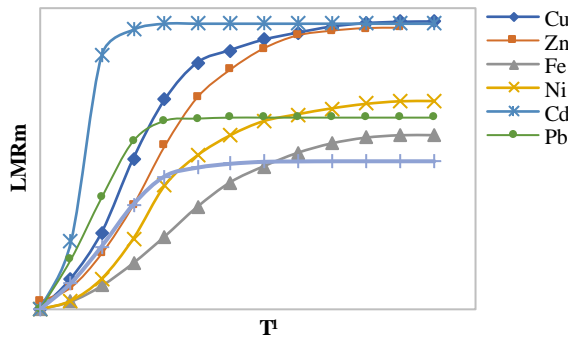


Figure 3. Elution curves (experimental) with diluted EDTA

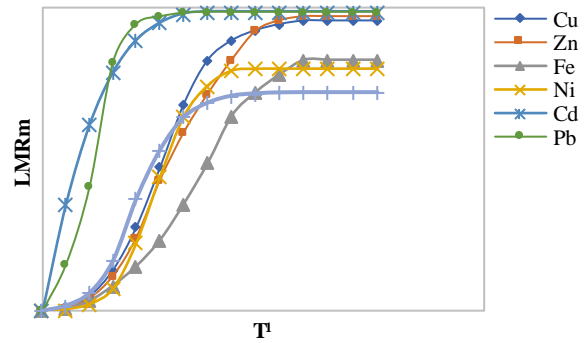


Figure 4. Elution curves (experimental) with diluted FeCl₃

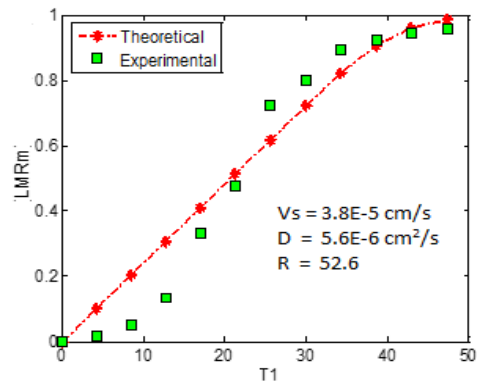


Figure 5. Elution curves of copper in soil

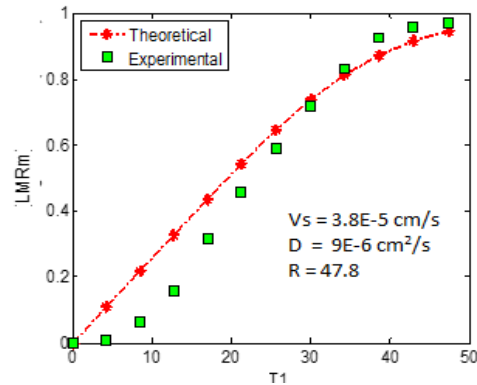


Figure 6. Elution curves of zinc in soil

TABLE 3. Transport rates of pollutants in soil

Pollutant	Dispersion coefficient (D) (cm ² /s)	Retardation Factor (R)
Copper (Cu)	5.6 x 10 ⁻⁶	52.6
Zinc (Zn)	9.0 x 10 ⁻⁶	47.8
Iron (Fe)	1.9 x 10 ⁻⁷	67.7
Chromium (Cr)	1.1 x 10 ⁻⁶	59.2
Cadmium (Cd)	7.0 x 10 ⁻⁵	22.6
Nickel (Ni)	2.8 x 10 ⁻⁶	62.8
Lead (Pb)	3.7 x 10 ⁻⁵	28.4

3.2. Batch Tests and Leaching Tests on Amended Soil Samples

The retardation factors estimated from the batch tests on amended soil samples with respect to the additives used are given in Table 4. The immobilization efficiencies of the additives estimated through the results of leaching tests are summarized in Table 5. From these tables, it can be observed that the immobilization efficiencies of the additives are directly proportional to the retardation factors obtained from the batch tests. Hence it can be concluded that the retardation factors play an important role in selecting the suitable amendment for immobilization remediation. The immobilization efficiencies of metals with lime addition were higher than the other two additives. These results

were similar to the data reported by Salihoglu [46]. The leachability of metals from the amended mixtures of this study were in the following order.

Cadmium> Lead> Iron> Nickel> Zinc> Copper> Chromium

The standard sequences of solubility of metal hydroxides with pH value are in the following order [47].
Cd(OH)₂> Pb(OH)₂> Zn(OH)₂> Cr(OH)₃> Fe(OH)₂> Ni(OH)₂> Cu(OH)₂

The leachability orders of metals from this study were observed to be similar to these sequences with small variations. These variations may be due to the solid matrix created by cementing agents and cation exchange with soil.

TABLE 4. Results of batch tests on amended soil

Amendment	Retardation factors						
	Copper	Zinc	Iron	Nickel	Cadmium	Lead	Chromium
Lime	58.3	56.2	76.2	68.2	29.8	32.1	69.9
NaOH	54.8	52.4	69.6	63.8	23.9	29.8	61.2
Cement	55.7	55.8	71.3	64.4	25.6	30.9	64.7

TABLE 5. Results of leaching tests on amended soil

Amendment	% leached and % efficiency	Metals in soil						
		Copper	Zinc	Iron	Nickel	Cadmium	Lead	Chromium
Lime	% Leached	22.6	24.5	36.8	31.2	44.1	43.4	17.5
	% Efficiency	77.4	75.5	63.2	68.8	55.9	56.6	82.5
NaOH	% Leached	39.8	38.6	41.9	43.7	51.5	49.8	29.6
	% Efficiency	60.2	61.4	58.1	56.3	48.5	50.2	70.4
Cement	% Leached	31.6	32.5	39.8	38.1	47.5	44.2	20.1
	% Efficiency	68.4	67.5	60.2	61.9	52.5	55.8	79.9

4. CONCLUSIONS

The determination of transport parameters has been described through batch tests and column leaching tests. The usefulness of the transport parameters, thus determined is demonstrated in the design of soil remediation techniques such as soil washing and immobilization. The column leaching tests carried out with diluted acid solutions on a contaminated dump site soil were evaluated to assess the suitability of the chemical solutions for soil washing remediation. The contaminant transport parameters of the pollutants were estimated through elution curves and the importance of these parameters in selecting the type and quantity of leaching solution for soil washing was discussed. The immobilization efficiencies of three additives were studied corresponding to a pH value of 10. The

retardation factors and long-term efficiencies of the amended soil samples were estimated through batch tests and leaching tests respectively. The results of these tests showed that the retardation factors are directly related to the immobilization efficiencies. Hence it was concluded that the retardation factors are useful to select suitable amendment for soil remediation using immobilization technique.

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Persian Abstract

چکیده

اصلاح خاک در یک سایت آلوده نیاز به آگاهی از پارامترها و فرآیندهای انتقال آلاینده دارد. این مقاله تعیین پارامترهای حمل و نقل از آزمایش‌های آبشویی ستون را در زمینه با دو تکنیک اصلاح خاک یعنی شستشوی خاک و بی‌حرکتی ارائه می‌کند. برای ارزیابی تکنیک شستشوی خاک، آزمایش‌های آبشویی ستونی بر روی خاک آلوده با محلول‌های اسید رقیق شده اسید هیدروکلریک، اتیلن دی آمین تتراستیک اسید و کلرید آهن برای ارزیابی کارایی آبشویی محلول‌های شستشوی انتخابی انجام شد. نتایج بدست آمده نشان می‌دهد که بازده محلول کلرید آهن رقیق شده بیشتر بود، زیرا درصد بیشتری از فلزات را از خاک حذف می‌گردد. از این نتایج آزمایش، پارامترهای انتقال آلاینده یعنی ضریب تاخیر و ضریب پراکندگی تعیین شد که برای محاسبه حجم محلول شستشو که برای شستشوی خاک در یک سایت مورد نیاز، مفید است. به عنوان بخشی از مطالعه تثبیت بر روی این خاک، خاک با اصلاحات انتخابی (آهک، هیدروکسید سدیم و سیمان) مخلوط شد تا pH خاک به ۱۰ افزایش یابد و عوامل تاخیری از طریق نتایج آزمایش آبشویی ناپیوسته برآورد شد. فاکتورهای تاخیری فلزات مختلف به‌دست آمده با افزودن آهک بالاتر از سایر اصلاح‌ها بود. برای تجزیه و تحلیل پایداری طولانی‌مدت مخلوط‌های اصلاح‌شده، آزمایش‌های آبشویی روی نمونه‌های خاک اصلاح‌شده انجام شد و راندمان تثبیت تخمین زده شد. مشخص شد که راندمان تثبیت با افزودن آهک بیشتر بوده و همچنین نتیجه گرفته شد که راندمان تثبیت مستقیماً با عوامل عقب ماندگی مرتبط است.
