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Heavy lead (Pb) mobility in compacted clay soils under landfill liners

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Abstract

The movement of heavy metals and their concentration depth is an important aspect of landfill design. This paper presents the results of laboratory measurements of lead (Pb) mobility in compacted clay soils of landfill liners. The contaminant transport of the heavy lead metal through a compacted landfill barrier was modeled utilizing a 50 cm column leaching test. Test results gave clear indication that the highest surface concentration lead was measured in the top 20 cm layer suggesting that the clay liners should be constructed with at least 20-30 cm thickness with 98% compaction level. It is clear that as long as the porosity are maintained very low by the use of modified proctor procedure it is possible to maintain the heavy lead on the top 20 cm depth once the geomembrane layer fails. This study suggests that experimental sample height should be higher than 20 cm in contrast to conventional 10 cm thick samples.

Keywords: Lead concentration, heavy metal, pollution, landfill, proctor compaction

1. INTRODUCTION

Trace heavy metal concentration in clay soils at the bottom of the landfill is a major concern because of their threat to the drinking wells. Highly contaminated soils may occur at old and new landfill sites particularly those that accepted waste containing heavy metals. Preventing heavy metal pollution is critical because cleaning contaminated soils is extremely expensive due to its difficulty.

Heavy metal elements accumulate at the bottom of the old and even new landfills and can not be broken down like organic pollutants. Among the many heavy metals released cadmium, lead and mercury are of great concern to environment because of their high toxicity (European Commission, 1999[1]; European Environment Agency 2004, [2]),

Detailed studies by Nagaraj et al. 2001[3]; Islama et al. 2004[4]; Wu et al, 1998[5]; Phadungchewit, 1989 [2], Lu et al. (1985) [7]; Kjeldsen et al. [7], 2002; Øygard et al. [9], 2004; Guangxi Wu [10] examined and provided information of the movement of the heavy material trace. Early studies showed that although heavy metals tended to be leached out of fresh landfill, they later became largely immobilized Lu et al. (1985) [7]. However, recent studies by Kjeldsen et al. [8], 2002; Øygard et al. [9], 2004 have showed that 0.01% -0.02% of heavy metals in old landfills is leached out over 25 years.

Xian et al (2008) [11] monitored Cd, Cr, Cu, Ni, Pb and Zn heavy metal leachate in a boreactor landfill over 20 months period. At the initial landfill stage, the leachate exhibited high heavy metals release, high organic matter content (27000–43000 g l of TOC) and low pH (5–6).

The selectivity coefficients of cation exchange in literature is usually estimated as: Na<Kq<Cd<Zn<Mg<Cu<Ca<Pb. Farrah and Pickering (1977) [12] found the orders of the four heavy metals in clay as of: Zn<Cd<Cu<Pb for soil pH=3.5 to 6. Phadungchewit (1989) [6] reported the order as Cd<Zn<Cu<Pb for soil pH>3 for montmorillonite clay. It is clear that heavy metal Pb is the most critical pollutant heavy metal in terms of cation exchange.

Guangxi Wu (1998) [10] modeled the migration of the four heavy metals in a sand-bentonite clay column to simulate the transport through a landfill barrier. Numerical results showed that the mobility of the metals followed the order of Cu>Zn>Pb>Cd even tough the differences were relatively small. This conclusion is different than previous studies. Their study included 10 cm height column tests. Concentration profiles of the heavy metals exponentially dropped between 8-10 cm from the surface.

The Trabzon Province is located on north east part of Turkey on the Blacksea coast. 150 tons/day solid waste is disposed to the present landfill in Surmene town and the landfill volume has reached to its effective capacity. Two alternative landfill sites are searched and will be constructed before 2017. The municipal regulation requires at least 50 cm compacted clay liner built below the bottom of the landfill 50 cm thick HPDE geomembrane liners. The clay liner was placed in two lifts of 20 cm and was compacted with 15 tons dynamic roller compactor at least 3 passes. Water was applied prior to compaction of the liner at 5 percent optimum water content. The compaction procedure is almost equivalent to the modified proctor test procedure in terms of applied force per cm square.

Trabzon municipal traditional cleanup costs between $10.00 and $100.00 per cubic meter, whereas removal of contaminated material (*ex situ*) costs are as high $30.00 to $300/ m3. The compaction procedure for the clay liners are around $ 5 including transporting, laying and compacting the soil layer. The municipal also requires efficiency modeling to optimize leachate collection efficiency.

The residence close to the Surmene landfill located 50 km north east part of Trabzon city has complained that the smell was acidic. Moreover the wells water became odoured black liquid. The main reason for this particular problem was the tear of the geomembranes at the bottom of the landfill facility. Potential movement of metals was a main concern. However the measurements taken at the site indicated that there were no heavy metal concentration. One explanation was that the clay liner observed all the heavy metal particulars.

Objective

The movement of heavy metals and their concentration depth is very important for landfill clay liner design and construction. Determination of depth of lead (Pb) mobility in compacted clay soil under landfill liner in Trabzon was particular interest for the new designed municipal projects. Maintaining the lead concentration in 50 cm (by regulations) compacted thick clay layer is imposed by the municipal design procedures.

The main objective of this study was to determine the heavy metal lead (Pb) concentration depth by utilizing a large coulomb test. There are several standards detailing different laboratory sample studies, but they are not conclusive, because there are options (thickness, compaction procedure etc.) depending upon the use for which the sample data is planned. The layer thickness of the clay liner hence the test sample thickness is the most important factor that effects the leachate modelling. Considering that the leachate movement in vertical direction is more important than the horizontal, the sample height is particular importance. The soil samples height (or thickness) prepared according to above mentioned studies as well as other studies in literature varied between 5 to 40 cm. However, in reality the landfill barrier systems are compacted at least 30-50 cm thickness.

Literature studies found selectivity orders of the heavy metals in clay as of: Zn<Cd<Cu<Pb for soil with varying pH values. In this study, particularly uniform one type of clay soil was selected to minimize the parameters effecting the test results.

Also, the landfill compaction procedure requires standard or modified proctor compaction test before applying the same material on site. The literature studies has not agreed on one particular compaction procedure. Recent geotechnical studies have demonstrated that modified proctor represents the field conditions better than the standard tests.

2. METHOD

In this study a laboratory leaching test was carried out to investigate mobility of the lead contaminated compacted soil profile with 50 cm thickness and 25 cm diameter using plastic tubes. Almost all studies reported in the literature considered only up to 15 cm depth soil profiles and 5-10 cm diameter while neglecting their larger dimension interactions. The main objective was to determine the necessary thickness of the compacted clay liner to prevent any leakage of lead from geosynthetic liners just beneath the landfill. The experimental testing program included a series of leachate tests on compacted clay specimens.

In this study sampling from prepared cylinder samples were taken at every 5 cm depth. Small sample sizes were not taken, as this does not give information with respect to the spread of contamination through sample. For this proposed study 50 cm height and 15 cm in diameter samples were prepared using plastic tubes, as shown in Figure 1. The experimental setup included a 10 cm thick layer at the bottom mainly consisting of 19 mm gravels to provide similar landfill drainage conditions.

The soil sample used in the testing was pure CL clay classified in Unified Soil Classification USCS, system. The clay soils studied exists on 56% of state of landscape in Trabzon Province. It was generally dark yellow to brown in color.

Clay soils were transported to laboratory from the field and they were subjected to field moisture measurements by using TDR (Time Domain Reflectometry). Same measurement device was used in the laboratory prepared samples to be consistent with field conditions. The TDR devices was used to measure the water content variations 30 cm below the clay surfaces on the field. The device did not allow measurements below 30 cm.

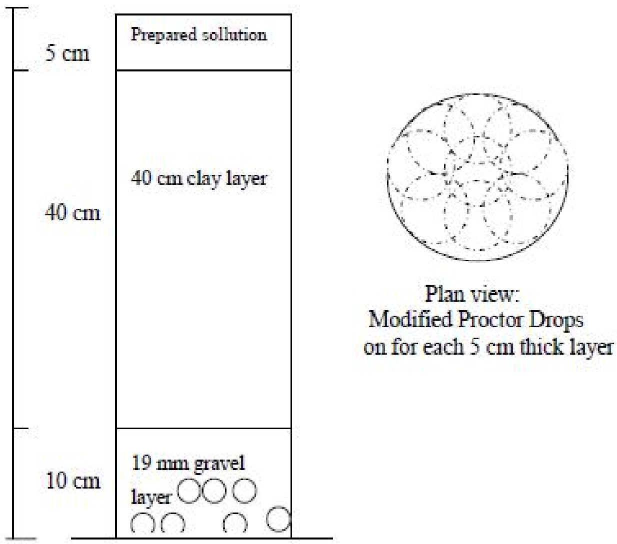

Figure 1. Profile and plan view of the cylindrical sample.

The prepared soil was unified Soil Classification of (CL) and contained a minimum of 50% by weight which passed 0.075 mm sieve. The soil had a clay content of 25% (less than or equal to 0.005 mm AS1152 sieve) by weight and had a plasticity index of 10 or greater. The classified soil had a hydraulic conductivity of not more than $1x10^{-7}$cm/sec. Soil compaction is required to have a relative compaction of 98% (modified proctor) and moisture content between optimum moisture content ± 2%.

Soil properties in the prepared sample did not vary from one layer depth to another since each layer was compacted with the same soil and compaction level at 25 proctor drops over 15 cm diameter sample, as shown in Figure 1. Small samples were taken from the center of the cylindrical compacted soil.

2.1. Soil Sample Preparation

This research consisted of performing modified proctor test (ASTM D 1557/AASHTO T 180) procedure on representative groups of Trabzon Province clay soils. The Department of Trabzon Municipal specifies use of modified proctor testing procedures to determine the compaction level of soils for roads and landfill construction projects. Recently, modified proctor test procedures have gained more acceptance since it is capable of achieving higher soil densities available from the compaction efforts. For the same soil, the optimum moisture content for a modified proctor test is usually less than optimum moisture content for a standard proctor test (ASTM D 698, AASHTO, T99) while maximum dry density is higher.

Clay soils were analyzed to determine particle size and Atterberg limits and subsequently classified into a Group Index. A sieve analysis was performed in accordance to AASHTO T87 (ASTM D 421). The Liquid Limit (LL), Plastic Limit (PL), Atterberg Limits were then measured for each soil in accordance to AASHTO T89 (ASTM D 4318). Five layers of soil were compacted in accordance to AASHTO T180 (ASTM D 1557). A small sample for each of the soils was taken to perform AASHTO T84 (ASTM C 128) procedures.

2.2. Sample Preparation for Measurement

The main objective of the study was to determine lead movement in the clay soil profile in a relation with concentration. For this purpose, 0.192, 0.479 and 0.720 g Pb(NO3)2 were used to prepare of 120, 300 and 450 ppm lead (II) nitrate solutions, respectively. The solutions were put into punched bags which were located on top of the compacted clay soil inside the 50 cm height and 15 cm diameter plastic tubes.

The clay soil was compacted according to the modified standard proctor procedure similar to the landfill clay liners. The experimental setups were separately subjected to for 4, 8, 12 and 16 weeks of time periods. A liter of 120, 300 and 450 ppm solution was given to each experimental setup every week. After providing lead exposure, 150 gr soil samples were taken at every 5 cm depth up to 45 cm.Total 108 soil samples (150 g, free of sand, gravel and plant materials) were collected and dried for a week at 40°C. Then, the samples were sieved and homogenized to a particle size of 63 μm and finally stored in polypropylene bottles at room temperature. A substrate material was required, thus 0.46 g of wax (high purity cellulose binder with 20 μm particle diameters) was pressed under 5 kN force, and then the mixture (0.48 g of specimen and 0.09 gof wax) was pressed together with substrate material with a compression of 10 kN. Samples for EDXRF analysis were usually prepared as powder pellets with the use of a binder. The useof powder pellets pressed without a binder eliminated sample dilution and therefore, increased the sensitivity and lowered detection limits (DLs) for trace elements. The other advantages of pressed-powder methods were its simplicity. Detection limit for this measurement was recorded as 0,9 mg/kg.

2.3. Analytical System

Pellets (solid) of 40 mm diameter with 1.2 g of 12 Certified Reference Materials (CRMs) with different elemental contents (but the same characteristics) were measured to calibrate the spectrometer for lead analyses. The measurement parameters were set up using the Epsilon 5 EDXRF system's (Epsilon 5 from PANalytical, Almelo, The Netherlands) in 1built software for lead analysis. Samples were irradiated by X-rays from a Gd tube under vacuum, equipped with a liquid nitrogen–cooled PAN-32 GeX-ray detector having a Bewindow thickness of 8 mm. The maximum power, typical current, and voltage of the instrument were set to 600 W, 6 mA, and 100 kV, respectively. For the measurement, Sr_Y_Pb_U condition and Mo secondary target were defined for 1000 second.

3. RESULTS AND DISCUSSION

Extracted small soil samples around 50 grams were collected from surface (0) to bottom (40 cm) of the prepared compacted soil at every 5 cm. Vertically, the content of lead concentration in soil decreased in two stages with respect to depth. The first stage was from the surface (0–20 cm depth), and the second stage was from the bottom horizon to the deeper horizon (20–40 cm depth). It was found that only the upper layer of soil (0-15 cm) contained lead. At least 80-90 % of activity remains in the upper 15 cm of soil.

Figure 2 shows the soil properties variation through the depth in relation to PPM data. The measured water by the moisture content device for same horizontal levels was identical indicating a homogenous compaction was obtained for the samples. This allowed to measure and interpret the transport of heavy metal in clay layer soils. The clay soil performed very well by absorbing the lead concentration in the first 15 cm. Thus, it was hard to mention if there was a correlation with concentration and void ratio the soil. Figure 2 shows the soil properties variation through the depth. The soil void ratio of samples ranged from 6 to 9, slightly above the optimum range (5.5-8.00) considered to be satisfactory for landfill clay liners. The available bulk density of the soil ranged from 1.6 to 2 t/m^3 which was in reasonable range for clay soils. The porosity values were below 0.5 indicating a good barrier for low permeability.

Figure 3 shows the concentration profiles of the lead heavy metal after 1, 2, and 3 months of solution staying on top of the sample. Soil pollution in the surface horizon showed similar trends for all samples subjected to 1, 2, and 3 month periods. Figure 3 shows that at the surface, total metal concentrations were exponentially distributed and decreased with increasing depth profile. The concentration of lead concentration away from the soil surface, around 20 cm, decreased exponentially with respect to that at the surface. The general trend line of the decrease can be formulated as follows:

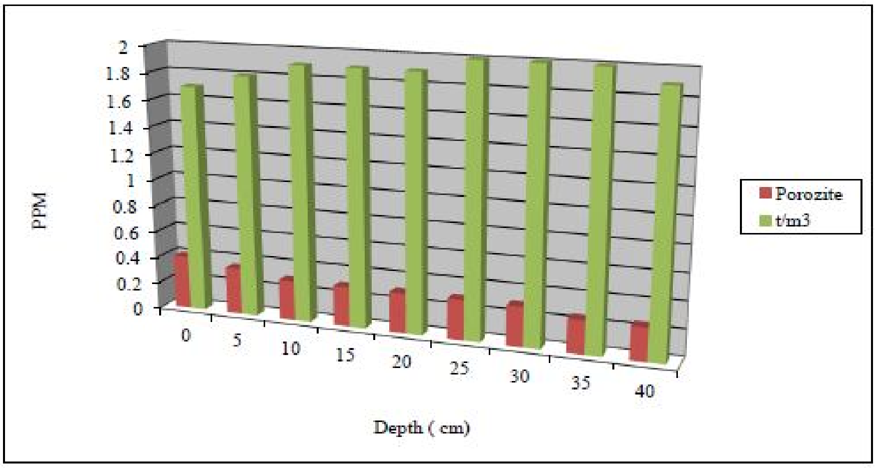

Figure 2. Soil properties variation through the depth in relation to PPM data.

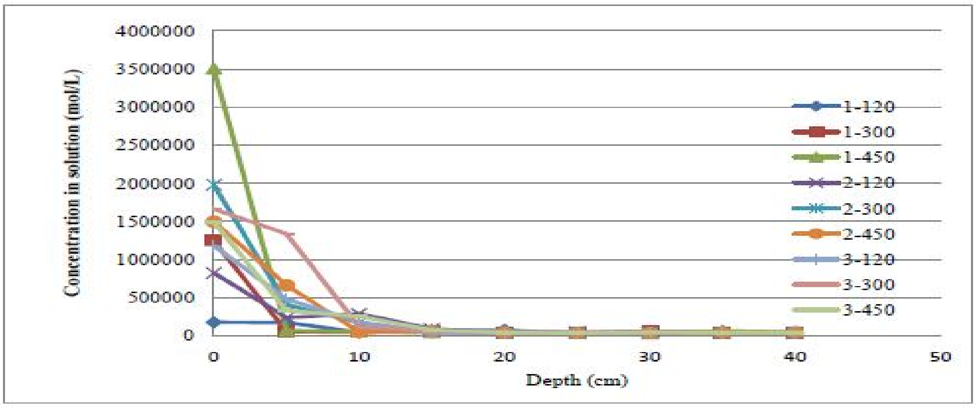

Figure 3. At the surface, concentrations were exponentially distributed and decreased with increasing depth profile.

Concentration (mol/L) = $4.3x10^5 x\ e^{-0.076x\ \textbf{Depth (cm)}}$ $R^2 = 0.8287$ (1)

Results also indicates that penetration of lead into the soil is a very slow process in this case. There were no correlation between the lead concentrations due to the fact that clay soil was compacted according to the modified proctor procedure and clay properties of the soil function very well as a barrier.

The symbols on Figure 3 indicates the solution time duration on top of the sample surface for one, two and three months. The following numbers shows the concentration in terms of ppm units. Findings revealed that an exponential relationship between the metal concentration and the depth from the soil surfaced. The graph findings are similar to the literature studies.

The content of lead concentration in soil decreased in two stages with respect to vertical depth. The highest surface concentration lead concentration was measured in the top 15 cm layer of soil profile and there was no detectable concentration below 15- 20 cm. Lead content was found from 1.6 to 2 ppm indicating a rather low range for most of the samples. It is clear that as long as the porosity are maintained very low by the use of modified proctor test procedure it is possible to maintain the lead on the top 15-20 cm depth.

Figure 3 shows the dissolved and exchangeable concentration profiles of lead (Pb2+) along the soil column. The concentration profiles are given for intermediate stages of one, two and three month days. It is observed from Figure 3 that (Pb2+) concentration in the soil solution along the column decreased creased with time, with the maximum concentration occurring at the top to the column where the leachate was supplied. It can be said that the lead concentration would not go further down 20 cm depth if the clay layer is compacted with 95 % compaction (modified proctor procedure) level and assuming that the base layer under the clay liner is stiff enough. Even though the results indicate that a 20 cm thickness layer would be okay to prevent lead leakage, it is suggested that a higher thickness layer should be compacted since the layer may not resist the shear forces above the layer.

During adsorption processes, pollutants are fixed at the surface of the adsorbents, separating them from liquid phase. Clay materials have already been shown to have high adsorption capacity, which may even exceed that of activated carbon under the same conditions of temperature and pH, because of their high specific surface area, high cation exchange capacity.

These species may increase the ionic strength of the soil solution, and may also compete with the heavy metal cations for the adsorption sites on the clay particle surfaces. The cause of this low depth penetration is the fixation of heavy metal by the crystal lattices of clay minerals. Clay minerals are leaf like structures composed of layered negatively loaded silicate platelets. In triple layered clay minerals cations are taken up between the layers to equalize ionic charges. Due to this specific binding, they are protected against leaching into deeper soil layers and are only limitedly available to plants.

The Trabzon Province municipal requires that liner efficiency modeling be performed to optimize leachate collection efficiency as well as to help predict the amount of penetration depth which will require treatment. The model developed by this study can be used by other states as a model, which assumes the existence of a uniform compacted liner and does not account for construction variability. With the use of the this model, the analysis showed that even with a uniform construction assumed, a linear thickness of less than 20 cm would result in a increase of leakage through the liner. As the liner thickness is increased, the flow through the liner is significantly decreased. The trend of decreasing flow is observed until a thickness of 20 cm is reached. It is clear that the sample heights below 20 cm may give over or under estimated results of leachate concentration. In this research samples were prepared according to civil engineering general accepted proctor test procedure which is the most widely used procedure around the world as well as in Turkey. Typical landfill clay liner design thickness require 50 cm thickness of clay liner may be an overestimation causing more cost. Also, compaction of 50 cm thickness of clay liner is difficult and the percent compaction may not be achieved on field. Thus, 30 cm clay liner is strongly suggested.

Recommendation

More studies and regulations is needed to minimize heavy metal mobility. The presence of organic substances, affects the mobility of heavy metals in clay soils, (Clemente and Bernal [13], 2006; Gondar et al., 2006 [14]; Kyziol et al. [15], 2006; Doig and Liber [16], 2007; Evangelou et al. [17], 2007). In this study, particularly humic organic substances were not used to minimize the complexity of the test and thus the data interpretation. There are plenty humic organic substances in the rivers and lakes of Trabzon province. Humic substances can be mixed with the clay materials before compaction process to either decrease the thickness or at least strongly prevent the leachate under the landfill.

4. CONCLUSION

The main objective was to determine the necessary thickness of the compacted clay liner to prevent any leakage of lead from geosynthetic liners just beneath the landfill. A model was developed based on the laboratory measurements to predict the amount of heavy metal penetration depth which will require treatment. The experimental testing program included a series of leachate tests on 50 cm height and 15 cm in diameter compacted clay specimens. Findings of this study revealed that a exponential relationship between the lead metal concentration and the depth existing compacted clay liner.

As the clay liner thickness is increased, the lead flow through the compacted clay liner is significantly decreased. The highest surface concentration lead was measured in the top 20 cm layer of soil profile and there was no detectable concentration below 20 cm. Column leaching tests gave a clear indication of the clay liners should be constructed with at least 20 cm thickness with minimum 95% compaction level. It is clear that as long as the porosity are maintained very low it is possible to maintain the lead on the top 15-20 cm depth. Experimental studies focused on this subject should have at least 20 cm height samples in contrast to conventional 10 cm height samples.

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