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Assessment of Heavy Metals from Landfill Leachate Contaminated to Soil: A Case Study of Kham Bon Landfill, Khon Kaen Province, NE Thailand

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Abstract: The distribution of heavy metals in landfill leachate contaminated to soil was investigated. Soil samples were collected at different times of the year as well as at various locations and depths in and around the landfill and throughout the contaminated area. The physical and chemical properties of the samples were analyzed. The results indicated that the heavy metals, namely Cd, Cr, Pb, Cu and Zn were significant concentrations in the soil within a radius of 2,000 m from the landfill. The Spearman's rank correlation coefficient indicated that the appearance of high Cation exchange capacity, clay content and organic matter are statistically correlated with the high heavy metals accumulation. Moreover, the Fe and Mn oxide/oxyhydroxides plays important role in controlling heavy metals sink in soil as pointed out by the Spearman 's rank correlation coefficient which corresponding to the soil type, red loess with high iron oxide, in the study area.

Key words: Soil contamination, heavy metals contamination, municipal waste, hazardous waste, opened dump site, red loess

INTRODUCTION

Sanitary landfill is the most common method of disposal of solid waste and the most popular ultimate disposal option (Ward *et al*., 2005). The disposed of municipal waste contain a mixture of many chemical compounds originating from the various discarded products (Jones-Lee and Lee, 1993; Slack *et al*., 2005). A number of these chemicals are released during the lifetime of the landfill and resulted in released of heavy metals to the environment. Consequently, an unknown number of toxic substances will be presented in the landfill leachate (Baun *et al*., 2004). Even though landfill technologies of engineered facilities are designed to eliminate or minimize the potential adverse impact of the waste on the surrounding environment, generation of contaminated leachate still remains an inevitable. The subsequent migration of leachate away from landfill boundaries and its release into the adjacent environment is a serious environmental pollution concern (Jones-Lee and Lee, 1993).

Leachate migration has been implicated worldwide in environmental pollution because it contains various types of toxic chemicals, for example organic and inorganic substances especially; heavy metals (Barker *et al*., 1988; Fetter, 2001; Voegelin *et al*., 2003; Slack *et al*., 2005; Sparks, 2005). Local soil, surface water and groundwater can be contaminated with the chemical substances from the leachate. This contamination could affect any adjacent environmental receptors such as vegetation, plants, soil

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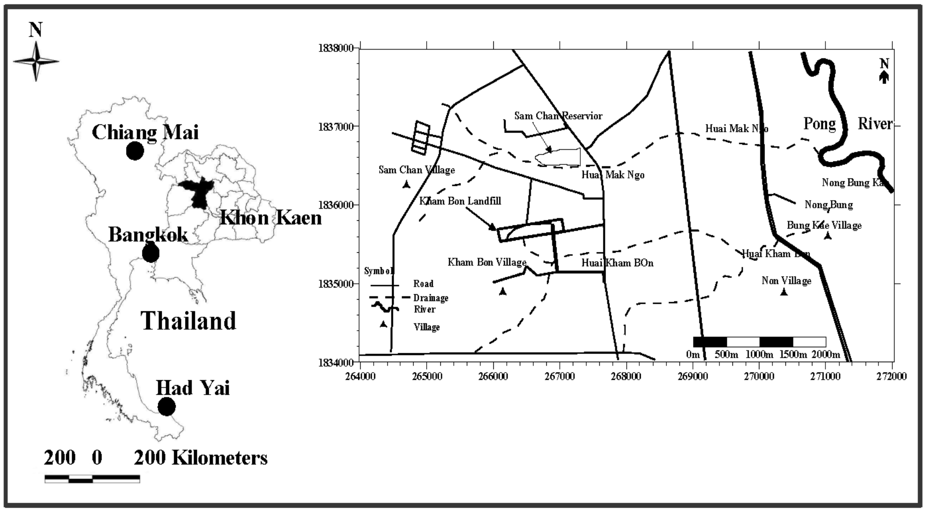

Fig. 1: The location of the study area at Kham Bon village, Khon Kaen Province, Northeast Thailand

as well as water supply sources, causing a risk to human health. Thus, it is important to understand the effect of leachate contamination on these receptors.

Heavy metals pollution has raised serious environmental concerns worldwide because bio-accumulation of these elements beyond the tolerance thresholds of living organisms pose long term risk to the earth's ecosystem (Voegelin *et al*., 2003; Sparks, 2005). The main flows of heavy metals to the environment are from industrial and municipal wastes, both of which contained a variety of toxic heavy metals. The heavy metals commonly found in landfill leachate include Cr, Cd, Pb, Hg, Ni, Cu, Zn, Fe and Se (Urase *et al*., 1997). The actual number and concentration of heavy metals in the leachate varies from one landfill to another.

Heavy metals in leachate from landfills have been extensively studied and monitored (Yong, 2001; Selim and Sparks, 2001). The major part of the metals is retained in the landfill. As a consequence, it must be expected that leaching of heavy metals from the landfills will continue for a long time (Freeze and Cherry, 1979; Fetter, 2001; Selim and Sparks, 2001; Yong, 2001). It can take years before groundwater pollution reveals itself and chemicals in the leachates often react synergistically and often in unanticipated ways to affect the ecosystem (Lee and Sheehan, 1996). Sharma and Reddy (2004) reported that waste containment and remediation problems require an understanding of the physical and chemical characteristics of the subsurface and the ability to engineer pollution control and remove the contaminants. Thus, it is important to characterize the site before starting others application.

The study aims to evaluate the vertical distribution and the downward mobility of Cd, Pb, Cr, Cu and Zn in soil around the landfill site as well as to determine the factors influencing mobility.

Description of the study area: The landfill site is located at Kham Bon village, Muang District, Khon Kaen Province, N.E. Thailand (Fig. 1). It is about 17 km North of Khon Kaen City along the Friendship Highway and comprises an area of 15 ha. The study area extends over some 3200 ha. The landfill is located on a ridge about 190 m above mean sea level at the interfluves between the Huai (rivulet) Mak Ngo to the north and the Huai (rivulet) Kham Bon to the South. The ridge is part of the rolling terrain of the high terrace, a geomorphological feature of the Khon Kaen region (Boonsener, 1991) and slopes gradually eastwards to the floodplain of the Pong River. The outskirts of these villages are used for growing rice, cassava and sugar cane as well as fish farming.

Chuangcham *et al*. (2005) reported that geologically the landfill site is underlain by fractured, reddish-orange siltstone of the Khok Kruat Formation which is part of the Khorat Group. The bedrock is overlain by silty sand soil attributed to the Red Loess lithostratigraphic unit of the Quaternary deposits of Northeast Thailand (Boonsener, 1991). These red beds form the foundation of the landfill. Red loess is composed of silt and fine to very fine sand. It is homogeneous and has no internal structure with quartz as the predominant mineral component. It also has very high dry strength but loses grain cohesion rapidly when it is wet. With reference to the collapsible behaviour of this loess, it can be interpreted that in rainy season, the leachate can flow through this loess more easily than in the dry season. Generally, the red loess layer varies in thickness from 1-8 m. The average thickness is about 5 m. It is situated approximately 180-220 m above mean sea level. From the borehole exploration, it is found that the average thickness of the landfill is 5 m.

The average annual rainfall is approximately 1750 mm, with about 80% falling between June and September. The average relative humidity is 75% and the potential evaporation rate is about 1575 mm year^{-1}, with the highest evaporation occurring between February and June. The average annual temperature is 26.5°C, with an average maximum of 33°C in April and an average minimum of 20°C in January.

Results from the aerial photograph interpretation and the topographic map analysis (Chuangcham *et al*., 2005) confirm that the topography of the study area plays an important role to the surface water flow directions at the landfill site. Water drains northwards from the landfill into the Sam Chan reservoir and then flows eastwards to the Huai Mak Ngo before discharging to the Pong River. Water draining southwards from the landfill enters the Huai Kham Bon and flows eastwards into an oxbow lake, Nong Bung on the Pong River floodplain. Field investigation revealed that the water table is located at 1 to 5 m below the ground surface in the shallow aquifer and is encountered from 9-15 m in the deep aquifer.

The Kham Bon landfill has been receiving refuse since 1968. Several of the old non-active refuse areas do not have liner systems installed. A loessial soil was used as daily and intermediate cover at the site. The site is capable of accommodating approximately 200 t day^{-1} of mixed wastes. The wastes which are disposed of at this site consist mainly of food and fruit, plastics, paper, wood, glass, metals and related municipal garbage. Additionally, hazardous wastes, for example, batteries, fluorescent lamp tubes and aerosol spray cans, are part of the waste stream (Kirathithorn, 2004). The proportion of different wastes at the Kham Bon landfill site is shown in Fig. 2 (Piyaprasit, 1996). Field observations indicate that the efficiency of the mixed waste treatment is likely to be poor. The landfill receives all types of waste materials, particularly solid and hazardous wastes. The different wastes are commingled together without proper sorting and are piled on the ground for natural decomposition. Frequently, the wastes are incinerated on the open ground so as to reduce their quantities. By such inappropriate disposal methods, the wastes have created not only a serious environmental pollution problem but also a threat to public heath and safety. They are a source of houseflies and produce unpleasant odors. Moreover, they could distribute disease pathogens and generate contaminated leachate. This leachate contains various pollutants and toxic substances, especially heavy metals, which migrating, infiltrating and descending in the soil profile to contaminate the adjacent surface water and groundwater (Boonsener *et al*., 1994; Chuangcham *et al*., 2005).

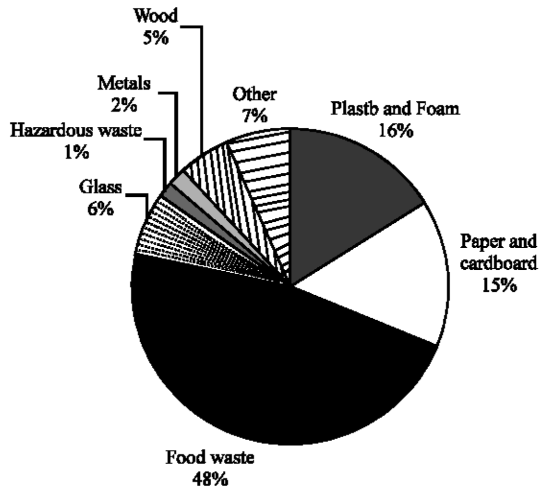

Fig. 2: Percentage distribution of wastes deposited in Kham Bon landfill (Piyaprasit, 1996)

MATERIALS AND METHODS

Soil sampling: A sampling strategy was established on the basis of site history and the preliminary reconnaissance to exhibit more variability in metal concentrations. Soils were sampled at different locations and depths in and around the landfill and throughout the contaminated parts of the site. The first sampling round was conducted on 12-13 December, 2004 during the dry, winter season. The second set of samples was collected on 14-15 May, 2005 during the peak dry period. The final set of samples were taken on 25-26 August, 2005 during the rainy season.

The aerial photograph and topographic map covered the study area were used planning for soil sampling. In the first sampling, 24 soil samples were gathered at a depth of 15 cm below the ground surface within a radius of 1500 m from the landfill and its surrounding area covering Kham Bon village (Fig. 3A). The results of sampling analysis were used to design the detailed sampling strategy.

Then, the second sampling was performed. The 38 soil samples were collected. Two samples were collected, one each at depths of 15 and 30 cm at each point from a random square grid on a circle within successive radii of 500, 1000, 1500 and 2000 m away from the landfill (Fig. 3B).

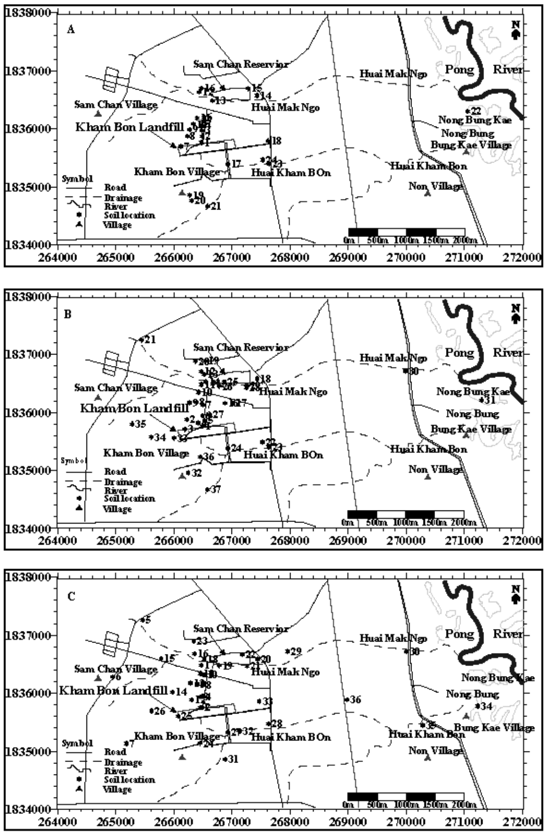

Fig. 3: Location of soil sample collected in all periods of sampling

The samples were closely spaced near the landfill site and along the ditch but spaced less closely with increasing distance from the site. Sam Chan village was included in this sampling to study the distribution of heavy metals falling within a radius of 2000 m.

The final sampling, 36 soil locations considerably polluted by leachate (Fig. 3C) was sampled at several depths. This final sampling was designed to obtain the spatial distribution of the contaminant concentrations both laterally and as a function of depth. The sample locations were selected from significant concentration of heavy metals revealed in the site as well as from the surface drainage in the study area. The samples were collected by hand auger from 3 depth intervals below the ground surface: 0-30, 30-60 and 60-90 cm. The zone of 0-30 cm depth is regarded as the disturbed zone I because within that range, farmers have used ploughs with small ploughshares (≈ 20-25 cm long) for cultivating crops whereas the zone of 30-60 cm is regarded as the disturbed zone II. This is because farmers have used ploughs with big ploughshares (≈ 45-50 cm long) during cultivation. The depth below 60 cm was determined as the zone of natural or artificially undisturbed soil. These soil samples were analyzed as a composite sample in each depth interval.

Soil analysis: The soil samples collected from the landfill site and adjacent area were air-dried and ground to pass through a 2 mm stainless steel sieve to remove gravel and rock, then were homogenized and stored for subsequent analysis. Physical and chemical properties of the soil samples were determined: Particle Size Distribution (PSD); pH; Electrical Conductivity (EC); Organic Matter (OM) and Cation Exchange Capacity (CEC). The PSD was measured and classified using a standard US Department of Agriculture soil textural classification (USDA classification). The soil pH was found from a 1:1 H_2O dilution. EC was measured using a conductivity meter. OM was determined by the wet oxidation method of Walkley and Black. Finally, the CEC was estimated by 1 M NH_4OAC method. For metal analyses, total recovery concentrations for all metals were measured using hot plate digestion with nitric acid following method 3050 B (US EPA, 1995). All metals (Cd, Cr, Pb, Zn, Fe, Mn) were analyzed using Shimadzu AA 6501F Series Flame Atomic Absorption (FLAA) Spectrophotometer.

The statistic analysis was performed by using Spearman's rank correlation coefficient, in SPSS 15.0 for Windows Evaluation Version, to test for a significant association between variables. The correlation between soil properties (clay fraction, pH, OM, CEC) and seven metals (Cr, Cd, Cu, Pb, Zn, Fe and Mn) were calculated. The contour maps were generated by surfer 7.0 from Golden software.

RESULTS AND DISCUSSION

Physicochemical characteristics of soils: The 108 soil samples were subjected to grain size analysis. The results revealed that the soils of the study area are medium to moderately fine-textured soils and include coarse-textured soil. They can be classified as Clay Loam (CL) to Silty Clay Loam (SCL), Sandy Loam (SL), Loamy Sand (LS) and Sand (S), respectively. The fine textured soil was found only in the eastern part of the study area (Nong Bung) which is the receptor of leachate from Huai Kham Bon. Focusing on the percentage of clay content distribution in the study area, the high distribution of clay content are found along the ditch of landfill leachate in the central, the leachate receptors; Huai Mak Ngo in the northern part, Huai Kham Bon in the southeastern part and Nong Bung in the eastern part of the study area (Fig. 4). Moreover, an

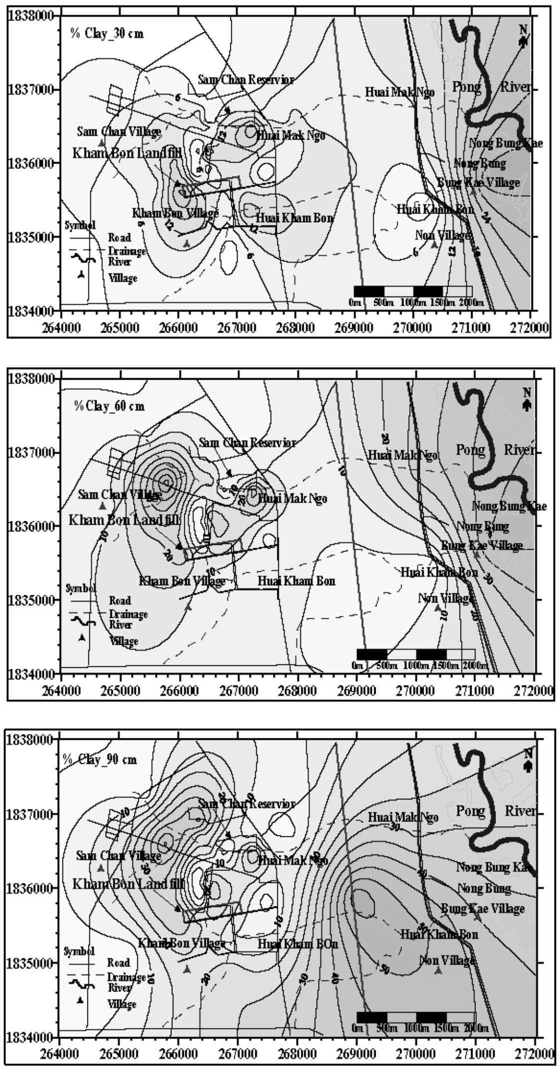

Fig. 4: Percent of clay content distribution for all soil at 0-90 cm depth in the third period of sampling

increasing of the percentage of clay content with depth was recorded in the soil collected throughout the study area. The medium textured soil was deposited in the central part of the study area which is near the leachate drainage ditch. The rest of the area is covered by the coarse textured soil.

Considering individual parameters, the pH values of 6-8.4 displayed relatively uniform distributions throughout the study area. Slightly acidic soil was found at the leachate ditch and near Sam Chan reservoir in the central to northern parts of the study area. In contrast, slightly alkaline soil was found in the eastern part of the study area.

The percentage of soil organic matter content ranged from 0.05-2.5%. Figure 5 indicates that the highest organic matter values were concentrated in the central and eastern parts of the study area. Moreover, it was observed that the organic matter are decreased descending with depth. This is because the top soil is composed of more humus which obtained from decomposed of plants and animals and the humus is decreased descending with depth (Sparks, 1986).

The cation exchange capacity of the soil samples ranged from 0.7-10.7 meq 100 g^{-1} of soil. As shown in Fig. 6 the soil samples collected in the eastern part of the study area displayed the higher CEC values for all different levels. Moreover, it was observed that the CEC value in the disturbed zone I is lower than other zones. The reason is that the disturbed zone I is the agricultural activities zone. It is always disturbed by human activities in all seasons resulting in the fluctuation of its properties. From the observation, it can be concluded that the high organic matter and clay content are related to the high CEC in soil (Banat *et al*., 2005; Sparks, 2005).

It should be remarked here that these physical and chemical properties of soil in the distributed zone I and II (0-60 cm depth) as reported above showed the fluctuation when comparing to the natural zone (60-90 cm depth) due to the agricultural activities.

Distribution of metals: Five metals Cr, Cd, Pb, Cu and Zn in soil samples were analyzed in this research as reported in Table 1. Heavy metals analysis from the first sampling period as shown in Fig. 7 pointed out that Cr concentration in soil varied within the range of 0.1-5.6 mg kg^{-1} whereas the concentration of Pb was in the range of 5-23 mg kg^{-1}. The Cd concentration in soil ranged from 1.8-3.0 mg kg^{-1}. Also, the concentration of Zn and Cu ranged from 0.9-55.0 and 0.9-12.7 mg kg^{-1}, respectively. The Zn and Cu concentrations are typical of landfill leachate. The sources of Zn are florescent tubes, batteries and variety of food wastes whereas discarded food is the main source of Cu. The Pb, Cd and Cr ions are toxic heavy metals (Roy *et al*., 1991) which are found in the study area. High levels of heavy metals were encountered in the Sam Chan reservoir and along the rivulet in the southeastern part of the study area. The highest concentration of heavy metals was recorded close to the landfill site within a radius of 500 m, decreasing with distance from the site. Focusing on the physicochemical parameters of soil that influencing the heavy metals deposition, it shows that the high heavy metals accumulation was agreed with the high of clay content, CEC and organic matter.

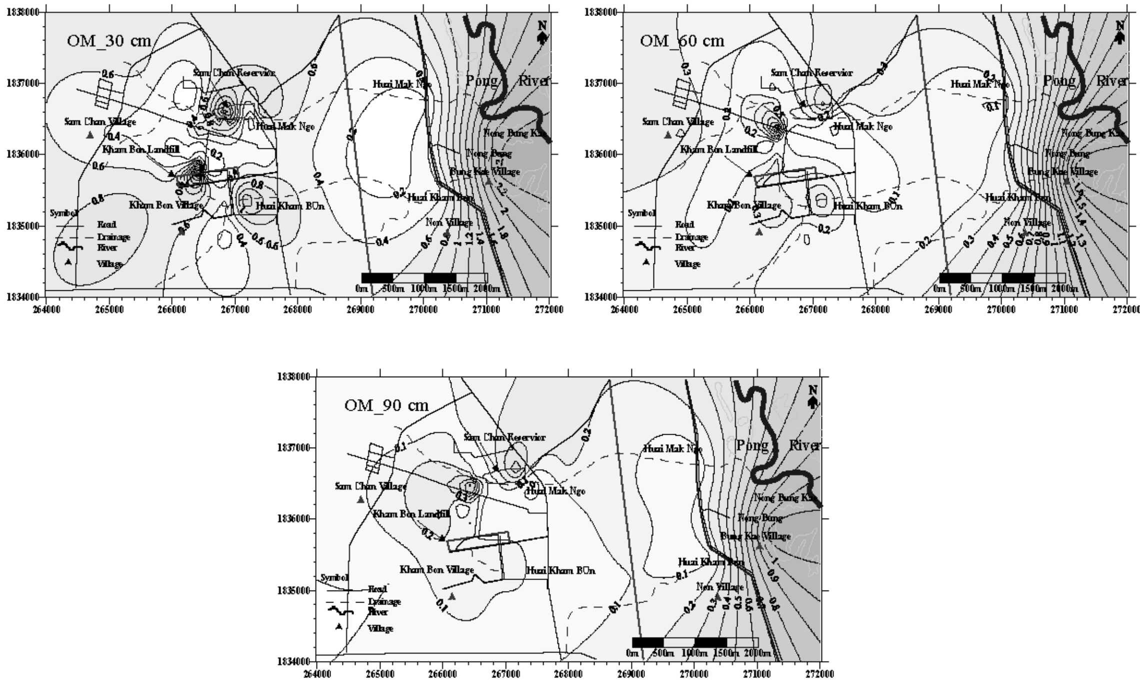

Fig. 5: Organic matter of all soil at 0-90 cm depth in the third period of sampling

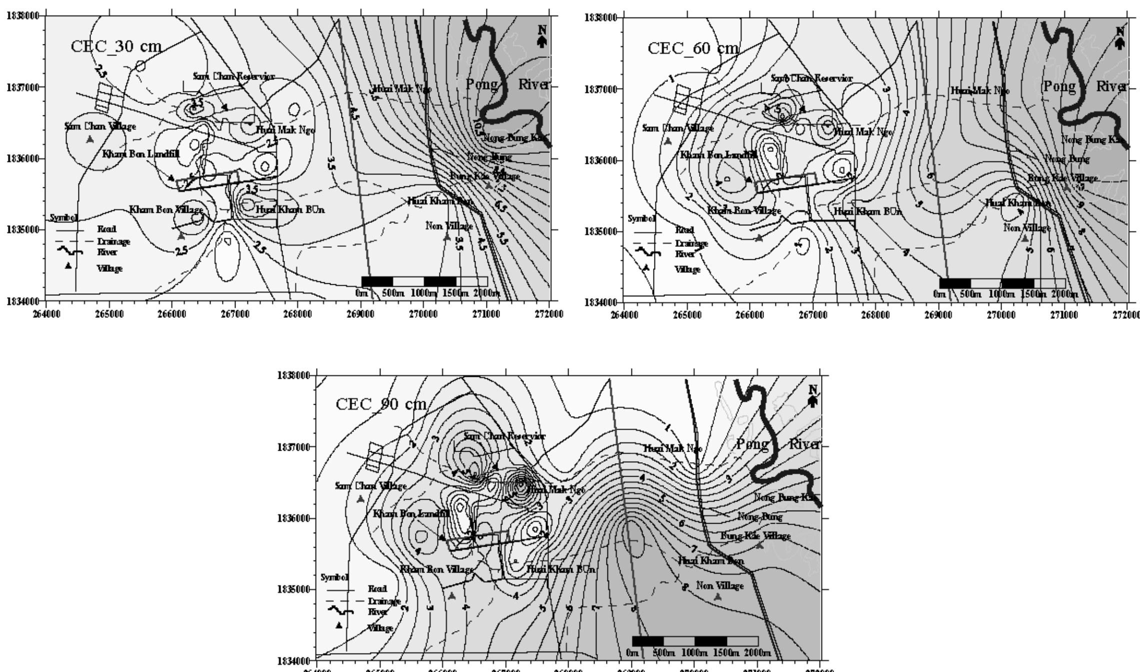

Fig. 6: Cation exchange capacity of all soil at 0-90 cm depth in the third period of sampling

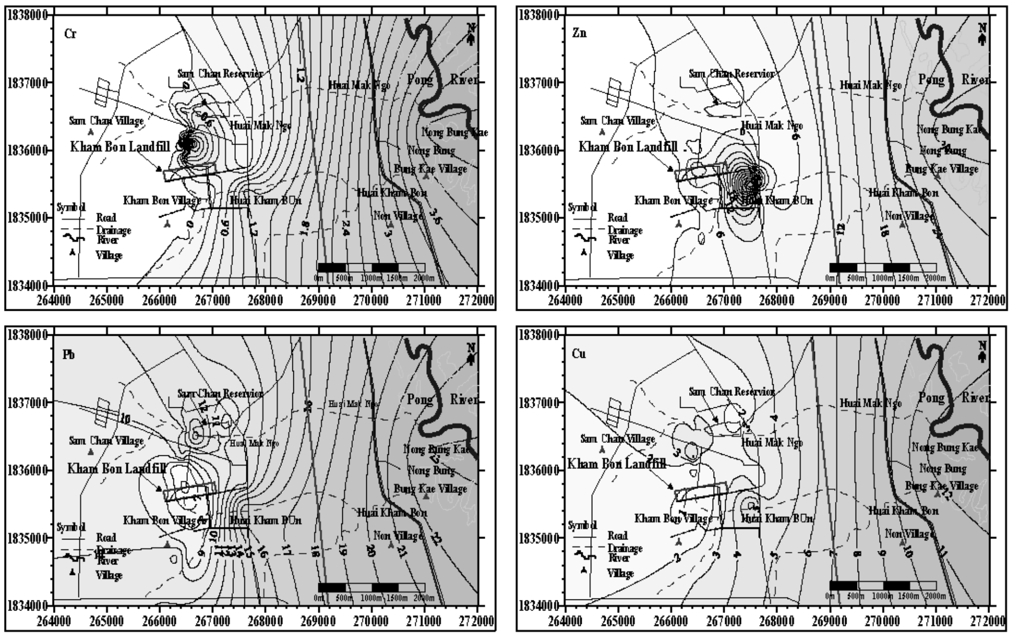

Fig. 7: Heavy metal concentrations at 0-15 cm depth in the first period of sampling

The study was extended in the second sampling period to collect soil samples at two levels (0-15 and 15-30 cm in depth) at each point of sampling (Fig. 8a). The concentration of Cr in the first depth interval, 0 to 15 cm below the ground surface varied from 0.14-14.99 mg kg^{-1}. Figure 8b expressed the Pb concentration which falling in the range of 2-43 mg kg^{-1}. The concentrations of Cu (Fig. 8c) and Zn (Fig. 8d) varied from 0.2-14.5 and 1-60 mg kg^{-1}, respectively. Within the second depth interval, 15-30 cm bgl, Cr concentration was in the range of 0.3-11.3 mg kg^{-1}, while the Pb concentration was between 6 and 42 mg kg^{-1}. The concentrations of Zn and Cu ranged from 0.2-80 and 0.1-33.8 mg kg^{-1}, respectively. It should be remarked here that, the concentration of Pb and Cr at the 15 and 30 cm depths, the Pb accumulated more at the 30 cm depth than at the 15 cm depth whereas Cr was deposited in the opposite manner. These phenomena are due to the great variation in pattern and contamination rates in depth as a result of pollutant discharge at the source. The fact that the lower strata seem to be more heavily polluted which may indicate that pollution at the source is decreased over the years, vice versa in the Cr.

Pb concentration (0 to 21.43 mg kg^{-1}) found in the third round of sampling was higher in the top 30 cm depth interval (Fig. 9a), decreasing with distance from the site. Possible sources of lead are batteries, chemical substances from photograph processing, lead-based paints and lead pipe deposited in the landfill. The high Pb concentration related to the high organic matter. In contrast to Pb, the Cr concentration (0-20 mg kg^{-1}) decreased with depth (Fig. 9b) which corresponding to the descending of clay content. Cd could not be detected in most parts of the surveyed area, except in some ditches and rivulets (0-1.70 mg kg^{-1}). The discarding of dry cell batteries and paint cans are possible sources of Cd. Pb and Cr values from all periods of sampling are at significant concentrations in the study area. This may depend on the amount of waste introduced each season in the landfill. The Cu (Fig. 9c) and Zn (Fig. 9d) contaminants of interest, varied from 0-40.4 and 0-52.2 mg kg^{-1}, respectively. The high Cu content related to the CEC and organic matter values. For the Zn value, the CEC, organic matter and clay content are influenced. The Fe (10-548 mg kg^{-1}) and Mn (3-3000 mg kg^{-1}) were also recorded in this period. They exhibited parallel distribution throughout the study area. During all three sampling periods it was found that the zone of highest concentration of heavy metals was the ditch of leachate which flows northwards to Sam Chan reservoir. Furthermore, other zones where the concentration is high are the downstream parts of Huai Mak Ngo and Huai Kham Bon where they discharge into the Pong River through oxbow lakes.

Moreover, the Pb concentration was found to be distributed both horizontally and vertically throughout

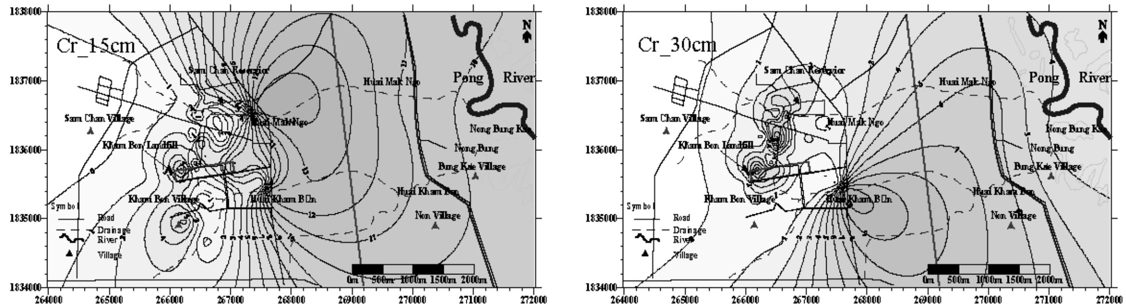

Fig. 8a: Heavy metal concentrations of Cr at 0-30 cm depth in the second period of sampling

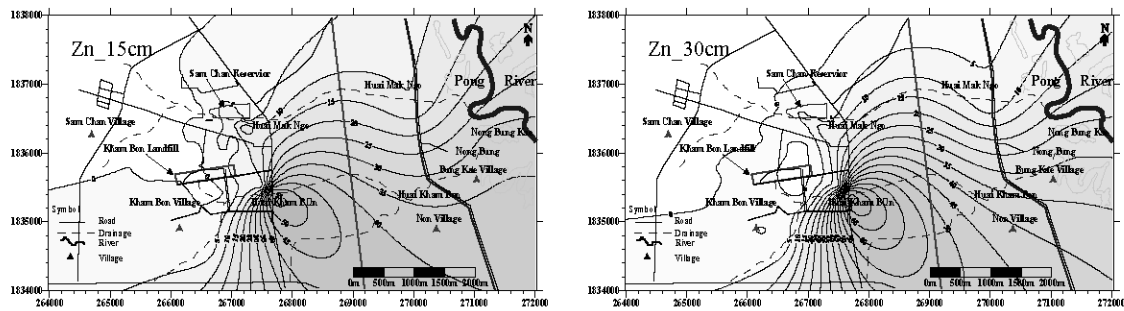

Fig. 8b: Heavy metal concentrations of Zn at 0-30 cm depth in the second period of sampling

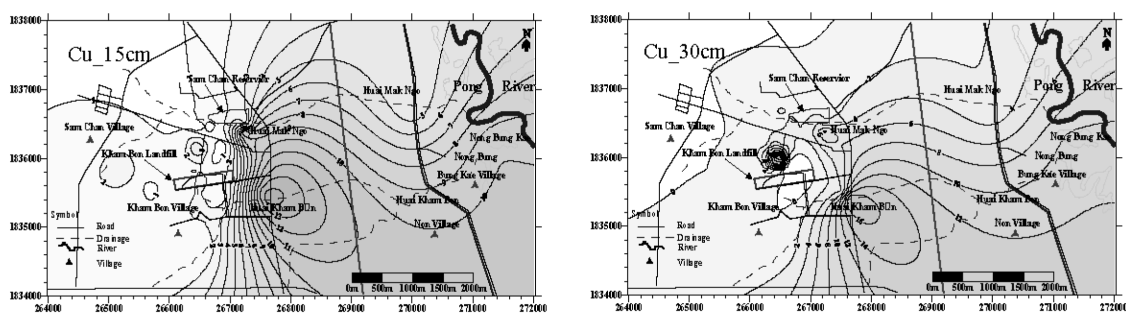

Fig. 8c: Heavy metal concentrations of Cu at 0-30 cm depth in the second period of sampling

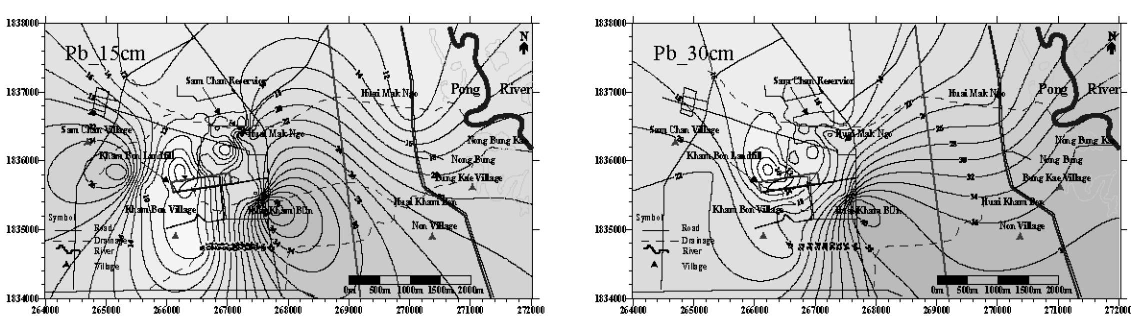

Fig. 8d: Heavy metal concentrations of Pb at 0-30 cm depth in the second period of sampling

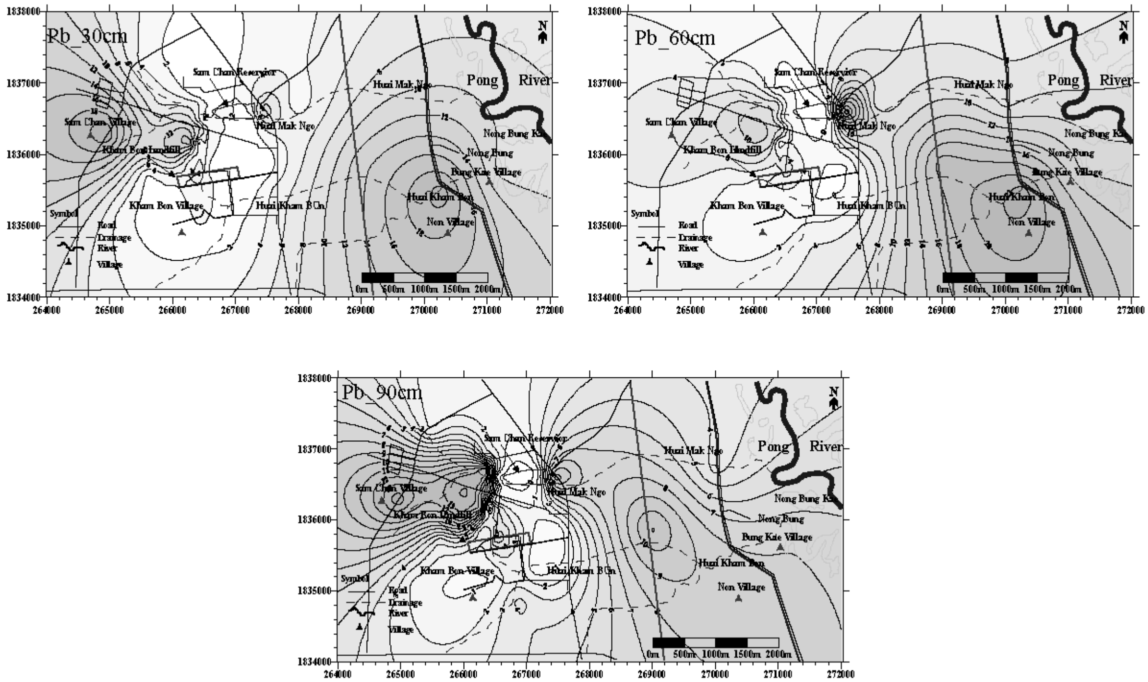

Fig. 9a: Heavy metal concentrations of Pb at 0-90 cm depth in the third period of sampling

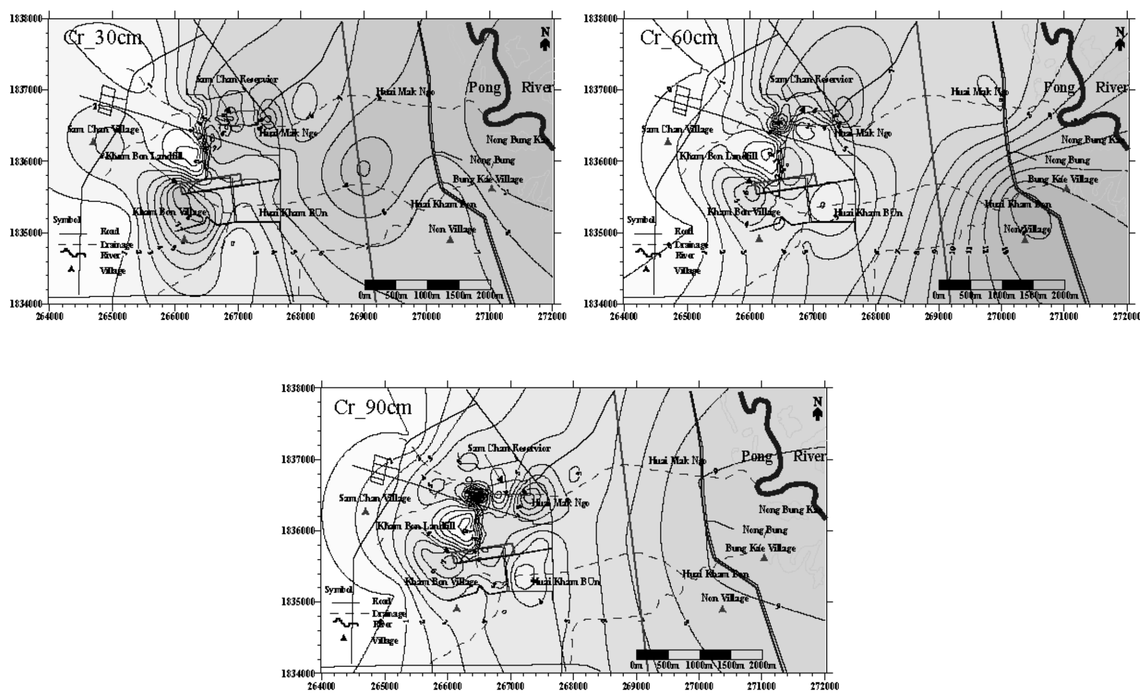

Fig. 9b: Heavy metal concentrations of Cr at 0-90 cm depth in the third period of sampling

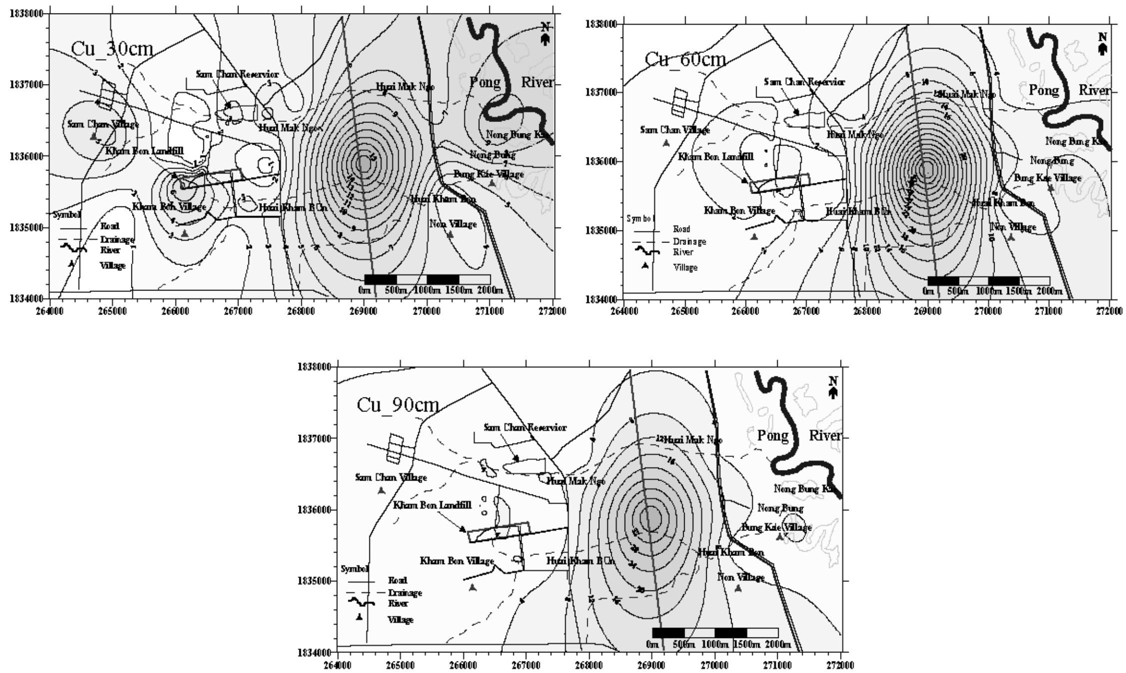

Fig. 9c: Heavy metal concentrations of Cu at 0-90 cm depth in the third period of sampling

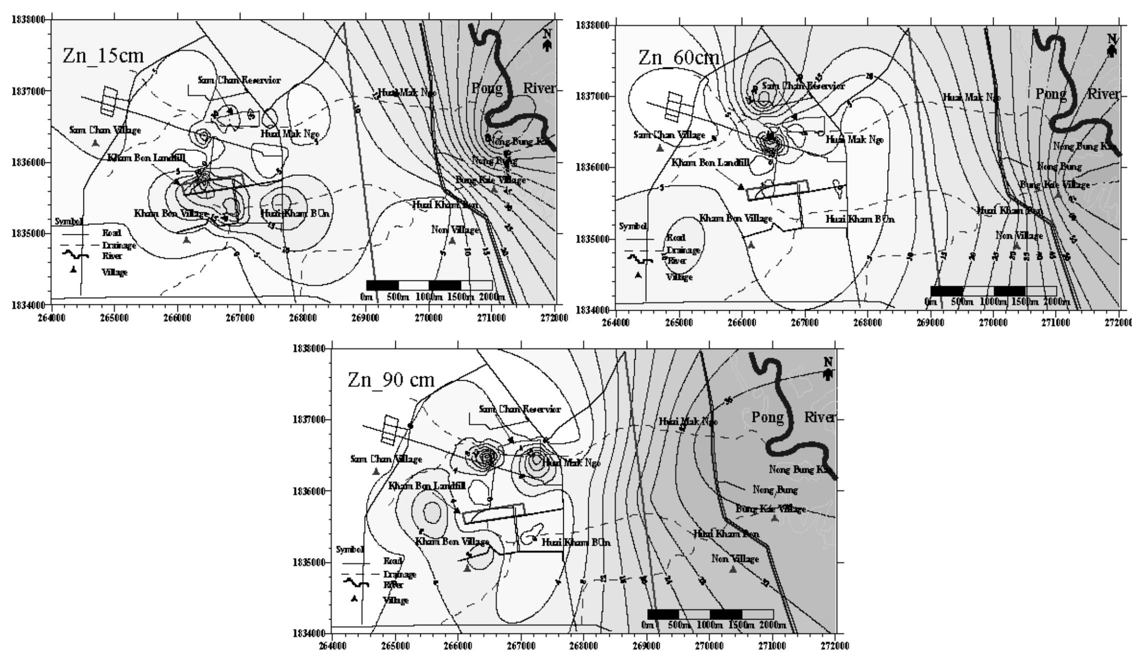

Fig. 9d: Heavy metal concentrations of Zn at 0-90 cm depth in he third period of sampling

the area. The field survey found that spray-paints cans, batteries, pesticide containers, iron pipes, dry cells and fluorescent tubes were widespread throughout the landfill. These mixed wastes had been burnt repeatedly in the open air within the site, in such a way that the heavy metals would also possibly be released by this action.

From the investigation in the second and third period of sampling, it was surprised that the significant Pb concentration was observed at Sam Chan Village in the northwestern part of the study area. In spite of the Sam Chan Village located approximately 200 m above mean sea level which higher than the Kham Bon landfill. Moreover, it is not situated in the direction of flow pattern as reported by Chuangcham *et al.* (2005). The reason of this appearance is due to the uncontrolled scavenger activities. They could be the heavy metals carriers.

Apart from physical and chemical properties controls, other factors of influencing heavy metals accumulation/ mobility were taken into consideration. The existence of red loess in the study area may be participated in the spreading of heavy metals. Moreover, the lower slope gradients at the lower lying creates lower-velocity flow, consequently, dominance of the finer fraction in the soil and higher retention of heavy metals was occurred as observed in the central part and eastern part of the study area. However, based on the Environmental Enhancement and Promotion Act B.E. 2535 (1992), the concentrations of cadmium, chromium and lead in soil do not exceed the standard allowable limit (Cd $\leq$ 37 mg kg^{-1}; Cr $\leq$ 300 mg kg^{-1}; Pb $\leq$ 400 mg kg^{-1}).

For the correlation analysis, data obtained from the third period of sampling time was calculated with the Spearman's rank correlation coefficient. The results illustrated that the measured heavy metals were positively correlated with soil properties. Considering in each heavy metal, The Zn exhibited the positively correlation with CEC (Spearman's rank correlation, $R = 0.629$, $p<0.0001$), clay content ($R = 0.606$, $p<0.0001$) and OM ($R = 0.474$, $p<0.004$). For the Cu, the positive correlation between Clay content ($R = 0.723$, $p<0.0001$) and CEC ($R = 0.671$, $p<0.0001$) was statistically significant. The similar patter was observed in Cr when considering with clay content ($R = 0.720$, $p<0.0001$) and CEC ($R = 0.590$, $p<0.0001$). The Pb showed slightly positive correlation with clay content ($R = 0.442$, $p<0.007$). A significant correlation was detected between the Fe and clay content ($R = 0.777$, $p<0.0001$) as well as the CEC ($R = 0.596$, $p<0.0001$). It is obvious that only the Mn correlated positively with pH ($R = 0.418$, $p<0.011$) and also correlated with CEC ($R = 0.574$, $p<0.0001$).

It is summarized that the most important factors influencing heavy metals accumulation in soils are clay mineral, CEC, metal oxides/oxyhydroxides (Fe and Mn oxide) and humic substance associated with natural organic matter (Bradl, 2004). In case of metal oxides and oxyhydroxides, the correlation of five heavy metals was calculated with the Fe and Mn. It was found that all heavy metals revealed the positive correlation with Fe ($R = 0.555$-0.777, $p<0.0001$) and Mn ($R = 0.385$-0.525, $p<0.0001$). Therefore, it can be stated that the metal oxides/oxyhydroxides plays important role in controlling the heavy metals deposition in the study area. Moreover, the study area is covered by the red loess which composed of more iron oxide (Boonsener, 1991) which is the source of sink for heavy metals.

CONCLUSION

Based on the experimental outcomes from both field and laboratory analyses, it is evident that soil at the Kham Bon landfill and its surroundings have been contaminated by leachate migration within a radius of 2000 m from the landfill. The soil analysis reveals that some heavy metals have accumulated at least 90 cm below the ground surface. Although the heavy metals seem to be scattered throughout the area without controlling factors, but there are patterns of distribution and controls. The first pattern is that the heavy metals distribution follows the drainage patterns within the area whereas the second pattern is controlled by the uncontrolled scavenger activities. They could be the heavy metals carriers. Other major factors controlling leachate production and migration at the landfill in this area are the seasonal variations in precipitation, the slope of the area which controls the runoff patterns and the soil type which affects infiltration and solute transportation to the water table. It is recommended to understand the long-term behavior of contaminants in the subsurface. This will require a proper understanding of the contaminant plume in the subsurface environment through knowledge of the sorption and transport properties of the soil and contaminants.

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