



## Soil Contamination from Non-sanitary Waste Landfill in Langat Water Catchment Area, Malaysia

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### Authors' contributions

This work was carried out in collaboration between all authors. Author SNSI designed the study, wrote the protocol, do the lab analysis and wrote the draft of the manuscript. Author CFI provide the lab facility for the sample analysis. Author MAAS managed the soil sampling. Author EMH managed the experimental process and the used of testing machine in the lab experiment while ASAW managed the literature searches and formatting of the manuscript. All authors read and approved the final manuscript.

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### ABSTRACT

Soil is a complex matrix and a major reservoir of contamination. It has the ability to bind many potential toxic elements such as heavy metals and they can exist in various forms. Leachate triggered soil contamination in waste landfill through sorption and migration process. This study determined heavy metals contamination in the soil of non-sanitary landfills in Langat river water catchment area in Selangor. Topsoil samples were collected from four landfill sites and three non-landfill sites for comparison. Soil type, soil pH, organic matter and exchangeable cations were

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determined using specified method. Available heavy metals in soil samples were determined using Ethylenediaminetetraacetic acid (EDTA). The distribution of heavy metals across landfill sites were performed using ArcGIS. Statistical Package for the Social Sciences (SPSS) statistical software was used to analyse the finding. The overall findings have indicated that Al (2340±575 mg/kg), Fe (2110±330 mg/kg), Cu (65.1±29.6 mg/kg) and Cd (4.99±1.3 mg/kg) were very high in the agriculture soil. Al and Fe also were high in the residential area soils with the mean ± SD of 2760±128 mg/kg and 1620±710 mg/kg respectively. Other elements such as Mn, Zn and Pb were highly detected in waste landfills. The highest Mn and Zn were detected in landfill site B with the mean ± SD of 76.5±13.9 mg/kg and 17.8±24.0 mg/kg respectively. High concentration of Pb was detected in landfill site D (37.9±37.8 mg/kg) and site B (37.5±24.3 mg/kg) while Cd was high in landfill site A (4.61±0.6 mg/kg) and site D (4.39±1.0 mg/kg). Ni was commonly detected as low in all sites. The highest Ni concentration was determined in landfill site B (1.55±3.06 mg/kg). In conclusion, all heavy metals in the studied soils were within the soil standard except for Cd and Cu. Most of heavy metals were retained at the boundary of the landfills. The mobility of heavy metals in the soil was possibly associated with the acidic-pH soils environment, high organic matter and clay content. In conclusion, the soil contamination occurred in both landfills and non-landfill sites where Cu contamination was found in agriculture soil, residential area and landfill site A while Cd contamination was present in all study sites.

**Keywords:** Soil; non-sanitary landfill; heavy metals; solid waste; langat catchment.

## 1. INTRODUCTION

Non-sanitary landfills and open dumping dominate the solid waste management system in most of developing countries [1,2]. Non-sanitary landfills are mainly operating in low and middle income countries as a result of limited technical and financial resources [3]. These landfills usually have lack of environmental abatement measures, without adequate protective facilities such as leachate treatment plants and lining materials on the bottom of landfills. This has inflicted a lot of contamination upon the environment especially to the soil.

Soil contaminations in landfill is triggered by leachate [4] through a series of chemical transportations by sorption and migration process [5]. There were 400 parameters and compounds which are more than 90 organic and metal organic compounds and 50 inorganic elements in the leachates [6]. The elements include halogenated aliphatic compounds, benzene and alkylated benzenes, phenol and alkylated phenols, ethoxylates, polycyclic aromatic compounds, Polychlorinated biphenyl (PCBs), chlorinated dioxins and chlorinated furans, pesticides, organic tin, methyl mercury and heavy metals [6]. The presence of a large number of hazardous compounds in leachate has obvious potential to contaminate the soils. Leachate is produced over time, and with the percolation of rain water, the degradable fractions of the waste decompose and the

resulting products are diluted and dispersed into the underlying soil if a site is not contained.

Previous research has indicated that the most common heavy metals detected in landfill soil were Cadmium (Cd), Copper (Cu), Iron (Fe), Zinc (Zn) and Nickel (Ni). For example, soil in the dumping ground and downslope of non-sanitary landfill in Dengkil, Selangor was detected with high concentration of Cu (2.21±1.17 mg/kg), Mn (11.35±1.30 mg/kg), Cr, (11.51±2.12 mg/kg), Ni (4.35±0.97 mg/kg), Zn (12.97±5.28 mg/kg), Pb (12.78±1.48 mg/kg) and Co (1.06±0.40 mg/kg) [7]. These elements were high within the near surface soil layer and decreased in concentration with increasing depth. It was due to the infiltration of leachate from the waste and the acidic pH soil environment influenced the contaminant dispersion in the soil [7].

Soil from a closed non-sanitary landfill in Kubang Badak, Selangor was determined with high Zn (257 – 666 mg/kg), Mn (29 – 263 mg/kg), V (4 – 175 mg/kg), Ga (34 – 182 mg/kg), Rb (64 – 136 mg/kg), Cr (38 – 137 mg/kg), Sr (0 – 159 mg/kg), Mg (16 – 174 mg/kg), Cu (0 – 202 mg/kg) and Pb (1 – 89 mg/kg) [8]. Soil from Kubang Badak landfill in Selangor was also detected with high As (0 – 27 mg/kg) which has exceeded the permissible concentration limits of Contaminated Land Exposure Assessment guideline (CLEA) (20 mg/kg) which was not suitable for land reclamation activities [8]. A study by [9] also has determined high concentration of As (64.4 mg/kg) and Hg (11.5 mg/kg) in soil sample from

a closed landfill in Kelana Jaya, Selangor. This was associated with the released of hydrogen sulphide from gypsum that was dumped in this area [9]. The industrial waste deposition into a non-sanitary landfill in Bukit Kemuning, Selangor has been associated with high concentration of heavy metals (i.e. Fe, Mn, Cu, Cr, Co, Ni, Zn and Pb) in the soil [10]. The industrial waste deposition is also has been associated with high levels of heavy metals in the leachate of a closed and post closure landfills in Selangor [11]. Percolation of leachate as surface runoff has caused contamination of the soil and groundwater with Sulphates (SO<sub>4</sub>), Nitrates (NO<sub>3</sub>), Nitrite (NO<sub>2</sub>) and Phosphate (PO<sub>4</sub>) [12].

Literature findings also indicate that heavy metals are probably derived from their soil parent materials such as crystalline schist (Cu) and igneous rock (Cd) [13]. They also could be generated from the application of phosphatic fertilisers [14,15], atmospheric deposition [16,17], fossil fuel combustion [18], refuse incineration, iron and steel production, sewage sludge [19], mining activities [20] and waste landfills [21,22].

This paper focuses mainly on the distribution of selected heavy metals on the topsoil of the non-sanitary landfill sites in the Langat river water catchment area, Selangor. This was performed by series of field and laboratory studies to determine the soil characteristics and heavy metals concentration, the level of heavy metals between sites, the relationship between heavy metals and soil properties and the distribution of heavy metals across landfills. This study was set within a broader assessment framework which is considered practical and applicable especially to developing countries in the Asian region.

## 2. MATERIALS AND METHODS

### 2.1 Description of Study Area

The study area is located at the Langat water catchment area in Selangor. The total area of the catchment is 1,815 km<sup>2</sup> and lies within latitude 2° 40' 15" N to 3°16' 15" and longitudes 101°19' 20" E to 102°1' 10" E. The main river course length is about 141 km mostly situated around 40 km east of Kuala Lumpur. The climate is wet and dry throughout the year. The temperatures range between 23°C to 33°C and the humidity levels generally 80%. The annual rainfall is generally more than 2,600 mm [23]. Langat is an important water catchment area providing raw water supply for the state. As a consequence of rapid

development, a significant environmental deterioration including soil contamination [24,25], water deficits and water quality deterioration [26] has become apparent. Several researchers have reported that the major source of soil pollution in the state was from waste landfills such as in [9,11,12,16,27,28,29,30]. Fig. 1 shows the location of the study area. There are in total 20 landfills of which 13 are closed and 7 still operating in the study area. Four of the landfills (3 operating and 1 closed) are located at in the Langat catchment area.

### 2.2 Soil Sampling

Topsoil samples (0 to 20 cm) were obtained from four non-sanitary waste landfills (Site A, B, C and D) and three non-landfill sites i.e. agriculture, industrial and residential area (Fig. 1). Soil samples from landfills were collected at five points in and around landfills (i.e. 0 m (immediate dumping area), 15 m, 30 m, 60 m and 100 m) while the sample for non-landfill sites were randomly taken. All of these sites are located on the main water catchment area of Langat basin. Samples were manually grabbed with a hand auger (Edelman augers) and were packed in plastic bags before being transported to the laboratory. Samples were air-dried at room temperature (22°C) and gently ground using a ceramic pestle and mortar. Soil samples were then passed through a 10 mm mesh sieve to separate the fine particles (< 2 mm) from coarser material. The soil samples of (< 2 mm) were then preserved in a sealed plastic bag for laboratory analysis [7].

### 2.3 Soil Laboratory Analyses

Soil pH was determined by pH meter (Metrohm) in a solution of aqueous soil suspension; 1 part soil to 2.5 parts solution. Total carbon was analysed with a LECO Carbon Analyzer CR-412, a non-dispersive, infrared, digital controlled instrument designed to measure the carbon content in a wide variety of organic materials including coal, oil and some inorganic materials including soil, cement and limestone. The total organic C was converted to organic matter by the Walkley-Black equation (Eq.1);

$$\begin{aligned} \text{Percentage of organic matter (\%)} &= \\ \text{Percentage of total C (\%)} \times 1.72 &\quad (\text{Eq.1}) \end{aligned}$$

The exchangeable cations were determined through leaching method and a flame atomic

absorption spectrophotometer was used to determine  $\text{Ca}^+$ ,  $\text{Mg}^+$  and  $\text{K}^+$ . Particle size of soils was measured by the pipette method. Heavy metals were determined with Ethylenediaminetetraacetic acid (EDTA). Samples were analysed for heavy metals by Inductively Coupled Plasma Spectroscopy (ICP-OES) (Model Iris Advantage ICP-OES Spectrometer). The detection limits for trace elements were 0.09  $\mu\text{g}/\text{ml}$  for Pb, 0.03  $\mu\text{g}/\text{ml}$  for Zn and Ni, 0.02  $\mu\text{g}/\text{ml}$  for Cd and Cu, 0.03  $\mu\text{g}/\text{ml}$  for Al, 0.005  $\mu\text{g}/\text{ml}$  for Fe and 0.002  $\mu\text{g}/\text{ml}$  for Mn. Precision of the metal measurement was determined by analyzing in triplicate the metal

concentrations. Results were analysed using the Statistical Package for the Social Sciences (SPSS) to obtain the level of contaminants and also to determine the correlation between parameters.

## 2.4 Spatial Interpolation with ArcGIS

GIS-based interpolation methods were used to generate spatial distribution maps in this study to determine the distribution pattern of heavy metals. Inverse Distance Weighting (IDW) method, with a power of two and radius of six

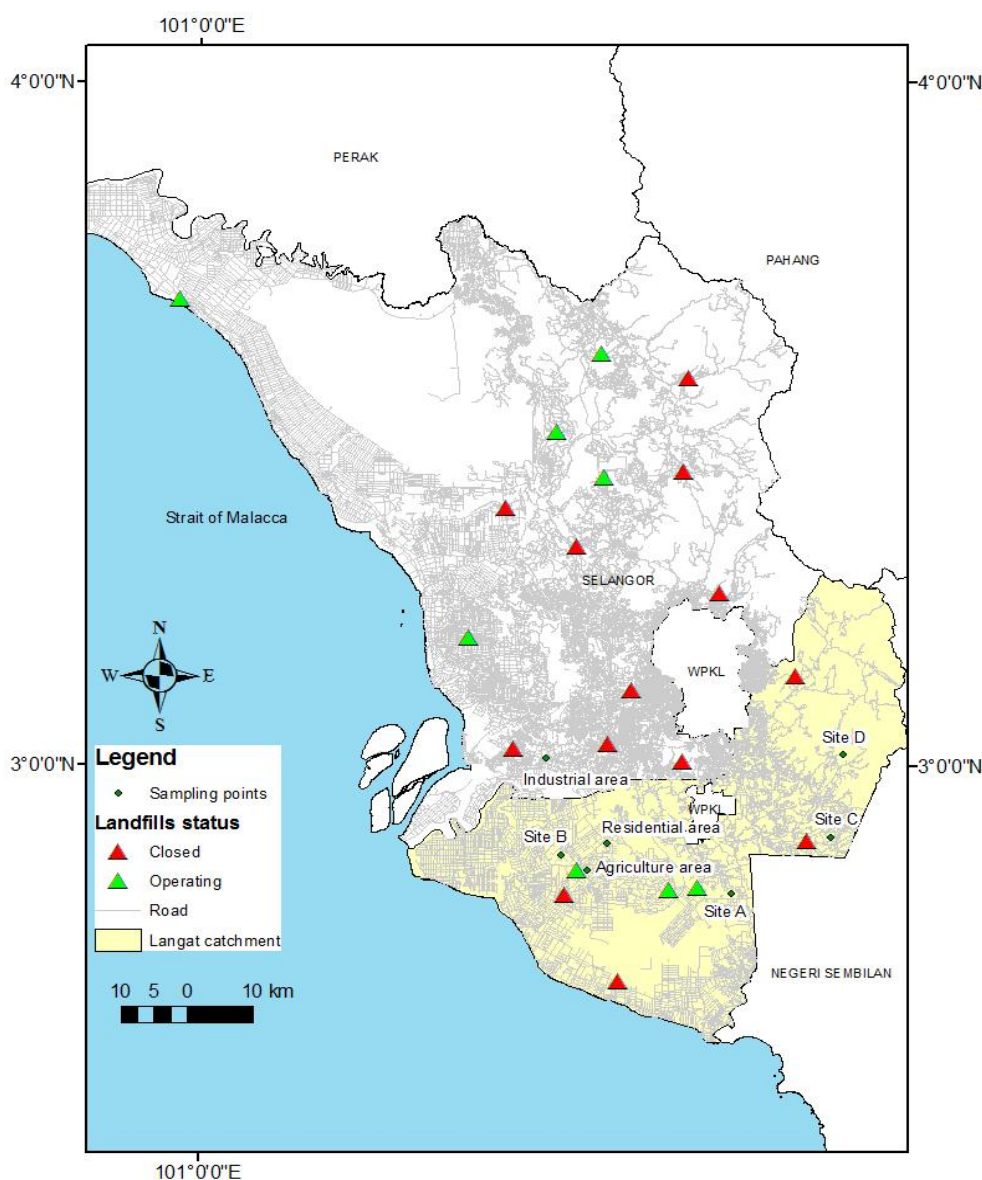


Fig. 1. The location of study area, landfill sites and sampling points

points appeared to be particularly appropriate in this study since it gave a smooth surface to the data and a root mean square error (RMSE) value that was sufficiently small for the purpose of the analysis.

Interpolation is essentially a means of estimating values for points where we have no measurements [31]. The goodness of interpolation can be characterized by the discrepancy of the interpolated value from the true one. Partly due to simplicity, IDW also suffers from several limitations, including the fact that the weighting parameters are chosen a priori, but not empirically determined [32]. In addition, IDW cannot estimate the variances of predicted values in un-sampled locations compared to what geostatistical methods such as kriging can provide [33]. Another limitation of IDW is that the distance decay parameter is applied uniformly throughout the entire study area without considering the distribution of data within it [32]. In other words, the standard application of IDW assumes that the distance-decay relationship is constant over space, though it may not be the case. A constant distance-decay value could be part of the reason that IDW provides less accurate predictions as compared to other interpolation methods [34,35].

### 3. RESULTS

#### 3.1 Soil Characteristics

The pH values of studied soils ranged from slightly acidic to highly alkaline between pH 3.01 to 5.11 (Table 1). Organic matter content was between 2.88% to 12.3%. The exchangeable cations were between 0.21 to 0.64 cmol/kg for  $K^+$ , 1.38 to 8.31 cmol/kg for  $Ca^+$  and 0.24 to 1.79 cmol/kg for  $Mg^+$ . These values are within the normal characteristic for Malaysian topsoil where the average pH values between 3.04 to 7.01, organic matter between 0.06 to 34.4% [14], the exchangeable cation for  $K^+$  was 0.27 to 3.31 cmol/kg,  $Ca^+$  between 0.34 to 21.8 cmol/kg and  $Mg^+$  between 0.72 to 2.04 cmol/kg [36][37]. There were two major types of soils in the region. The first being clay soils. This was the major soil type for site A, B and the non-landfill sites. The second type of soil is sand. This was the major soil type for site C and D. Clayey soils were slightly more acidic, has high organic content and exchangeable cations compared to the sandy soil.

#### 3.2 Trace Elements and Heavy Metals in the Topsoil

Table 2 shows the mean $\pm$ SD of trace elements and heavy metals in the topsoil. The most abundant trace elements found in the topsoil were Al (ranged from 389 to 2760 mg/kg) and Fe (30 to 2110 mg/kg). Al was high in residential and agricultural area with the mean $\pm$ SD of 2760 $\pm$ 128 mg/kg and 2340 $\pm$ 575 mg/kg respectively. Fe was high in agriculture and residential area with the mean $\pm$ SD of 2110 $\pm$ 330 mg/kg and 1620 $\pm$ 710 mg/kg respectively. Al and Fe were also abundant in landfill site A (1450 $\pm$ 820 mg/kg for Al and 930 $\pm$ 706 mg/kg for Fe) and site B (1290 $\pm$ 466 mg/kg for Al and 1260 $\pm$ 857 for Fe). Mn was high in landfill sites B (76.5 $\pm$ 13.9 mg/kg) and C (90.8 $\pm$ 30.7 mg/kg) while in other sites its remain low.

The highest concentration of Cu was detected in agriculture soil (65.1 $\pm$ 29.6 mg/kg). Cu also was detected in landfill site A (43.2 $\pm$ 34.5 mg/kg), residential area (42.9 $\pm$ 20.6 mg/kg) and site B (37.9 $\pm$ 23.5 mg/kg). Cu was detected as slightly low in landfill site C (20.5 $\pm$ 18.4 mg/kg), site D (13.7 $\pm$ 6.61 mg/kg) and industrial area (6.57 $\pm$ 3.43 mg/kg). High concentration of Pb was detected in landfill site D (37.9 $\pm$ 37.8 mg/kg), site B (37.5 $\pm$ 24.3 mg/kg) and residential area (27.9 $\pm$ 14.5 mg/kg). Pb in landfill site A, C and agriculture soil was slightly lower with the mean  $\pm$  SD of 20.1 $\pm$ 22.1 mg/kg, 17.9 $\pm$ 22.9 mg/kg and 14.9 $\pm$ 3.20 mg/kg respectively. Very low concentration of Pb was detected in industrial soil with the mean $\pm$ SD of 2.19 $\pm$ 0.69 mg/kg. High concentration of Zn was detected in landfill site B (17.8 $\pm$ 24.0 mg/kg), site D (14.8 $\pm$ 8.98 mg/kg) and site C (13.9 $\pm$ 16.1 mg/kg). The topsoil in industrial (8.82 $\pm$ 4.37 mg/kg), agriculture (6.24 $\pm$ 4.01 mg/kg) and residential area (4.06 $\pm$ 1.96 mg/kg) were detected with low Zn.

High concentration of Cd was detected in agriculture soils (4.99 $\pm$ 1.3 mg/kg), landfill site A (4.61 $\pm$ 0.6 mg/kg) and site D (4.39 $\pm$ 1.0 mg/kg). Cd also detected in industrial area (2.73 $\pm$ 2.62 mg/kg), site B (2.69 $\pm$ 2.13 mg/kg), site C (2.57 $\pm$ 3.11 mg/kg) and residential soil (1.6 $\pm$ 0.78 mg/kg). Ni was generally very low in the topsoil in this study ranged from 0.01 to 1.55 mg/kg. The highest Ni was detected in landfill site B with the mean $\pm$ SD of 1.55 $\pm$ 3.06 mg/kg. Other sites were detected with Ni of less than 1 mg/kg.

Table 1. Soil characteristics in this study

	Non-sanitary landfill sites				Non waste disposal site			Malaysian topsoil <sup>a</sup>
	Site A (n=31)	Site B (n=30)	Site C (n=34)	Site D (n=14)	Residential area (n=6)	Agricultural area (n=6)	Industrial area (n=6)	
Soil pH	4.87±1.20	4.55±0.78	5.11±0.85	4.95±0.42	3.01 ±0.37	3.44 ±0.57	3.94 ±0.56	3.0 to 7.0
OM (%)	12.3±7.16	5.78±2.40	3.91±1.80	2.88±1.43	7.61 ±2.10	8.50 ±3.74	5.88 ±3.16	0.03 to 34.4
Clay (%)	49.4±12.3	41.7±8.9	22.9±8.94	15.07±10.2	45.0 ±6.20	40.2 ±6.24	43.3 ±7.09	-
Silt (%)	35.9±8.88	38.7±7.09	17.7±8.39	19.3±12.3	40.0 ±4.15	40.7 ±4.32	36.0 ±7.27	-
Sand (%)	14.4±13.7	19.6±5.46	59.4±11.8	61.6±19.6	14.8 ±4.07	19.5 ±3.89	20.7 ±1.21	-
K+ (cmol/kg)	0.56±0.59	0.64±0.73	0.43±0.22	0.21±0.07	0.48 ±0.30	0.63 ±0.54	0.26 ±0.13	0.27 to 3.31
Ca+ (cmol/kg)	8.31±9.91	4.30±4.28	2.91±3.30	1.38±0.71	2.66 ±1.43	1.61 ±1.17	1.73 ±1.49	0.34 to 21.8
Mg+ (cmol/kg)	0.78±0.86	1.79±1.39	0.37±0.38	0.24±0.21	0.48 ±0.26	1.02 ±0.49	0.79 ±0.63	0.72 to 2.04

Note: OM = organic matter. Results expressed as mean ± SD. The number of samples for each site is indicated by n, <sup>a</sup> [14,36,37]

Table 2. Trace elements and heavy metals concentrations in the top soils (mg/kg)<sup>1</sup>

	Non-sanitary landfill sites				Residential (n=6)	Agricultural (n=6)	Industrial (n=6)	Target value (mg/kg) <sup>a</sup>	Intervention value (mg/kg) <sup>a</sup>
	Site A (n=31)	Site B (n=30)	Site C (n=34)	Site D (n=14)					
Al	1450±820	1290±466	782±534	389±252	2760±128	2340±575	683±179	NA	NA
Fe	930±706	1260±857	582±528	299±302	1620±710	2110±330	30.3±10.3	NA	NA
Mn	19.7±25.9	76.5±76.4	90.8±179	32.9±26.8	24.3±11.3	36.2±26.0	8.24±4.18	NA	NA
Cu	43.2±34.5	37.9±23.5	20.5±18.4	13.7±6.61	42.9±20.6	65.1±29.6	6.57±3.43	36	190
Pb	20.1±22.1	37.5±24.3	17.9±22.9	37.9±37.8	27.9±14.5	14.9±3.20	2.19±0.69	85	530
Zn	13.9±16.1	17.8±24.0	6.47±5.87	14.8±8.98	4.06±1.96	6.24±4.01	8.82±4.37	140	720
Cd	4.61±3.23	2.69±2.13	2.57±3.11	4.39±3.76	1.6±0.78	4.99±3.23	2.73±2.62	0.8	12
Ni	0.8±1.7	1.55±3.06	0.8±1.5	0.28±0.21	0.19±0.2	0.01±0.03	0.13±0.10	35	210

Note: <sup>1</sup> Trace elements in this study were Al, Fe and Mn. Heavy metals were Cu, Pb, Zn, Cd and Ni. Results expressed as mean ± SD mg/kg. The number of samples for each site is indicated by n. <sup>a</sup> The target and intervention values of Dutch soil protection / Malaysia soil standard (soil containing 10% organic matter and 25% of clay) [38]

The comparison of heavy metals in this study to the Malaysian soil standard which is adopted from Dutch Soil Protection Act [38] indicate Cu and Cd exceeded the Target value, but were not above the Intervention level and so were classified as slightly contaminated. Other elements i.e. Pb, Ni and Zn were classified as within the background values and not contaminated. The soil contamination occurred in both landfills and non-landfill sites where Cu contamination was found in agriculture soil, residential area and landfill site A while Cd contamination was present in all study sites.

### 3.3 Relationships between Elements and Soil Properties

Statistical analysis was performed to determine the relationship between elements in this study and the soil properties in Table 3. Al, Cu and Fe have positive relationship with clay, silt and organic matter and negative relationship with soil pH and sandy soil. This suggests that the concentration of elements is lower in soil solution of alkaline and neutral soil than in light acid soils. This indicates that Al, Fe and Cu have similar provenance to clay, silt and organic matter and tend to be concentrated in clay acidic soils rather than in alkaline sandy soils. This also explains why these were higher in the agriculture and landfill sites which located on the clay soil rather than in site C and D which located on the sandy soil. Mn and Ni were associated with  $Mg^+$  and  $Ca^+$ . Cd and Zn have positive relationship with organic matter although at low significant value. All elements have positive relationship with exchangeable cations except for Cd.

### 3.4 Spatial Distribution of Potential Toxic Elements (PTE)

Heavy metals in all landfill sites were interpolated and only the one with strong pattern in site A and B were illustrated in this paper. All elements tested i.e. Cu, Cd, Pb, and Zn was highly concentrated at the boundary of the landfills. None of these elements concentrated on the immediate dumping area (the centre point of the site) (Fig. 2).

## 4. DISCUSSION

Al and Fe in this study were high for agriculture soil, residential area, landfill site A and B. This was possibly related to the clay soil type in these areas. Al and Fe are the main components in clayey soil [39]. In more rigorous weathering

environments such as in tropics, these elements are abundant in clay minerals [39]. They also exist in the form of oxides that tie up toxic elements and influence their mobility [39,40].

Mn was high in landfills (site B and C). However, this element is within the normal concentration as Mn in rocks is reported within 350 to 2000 mg/kg [40]. High Mn in these sites was possibly associated with the pH of the soil. According to [39], Mn is mostly available in the soil with pH value of 5 to 6.5. These were the ranged of pH value for soil in site B and C. Besides, high organic matter content also associated with Mn concentration in the soil [39].

Cu in the agriculture soil in this study was higher than the average total content of Cu in Malaysia topsoil as reported in [14]. The average of Cu in the topsoil is reported as  $16.4 \pm 10.6$  mg/kg, four times lower than the value detected in this study. Cu in this study also higher than the cultivated soils in Peninsular Malaysia (43.8 mg/kg) as reported in [41] and in landfill soils (1.0 to 3.6 mg/kg) as reported in [12]. High Cu in the agriculture soil was possibly associated with the application of agricultural chemical-based products such as pesticide and fertiliser which has resulted in the concomitant increases of As, Cu, Cd and Zn [14]. In addition, the mixed of waste deposited including agricultural chemical-based products on the landfills [40] probably cause high Cu detected in landfill site A and B.

Pb in this study was high in landfill site D ( $37.9 \pm 37.8$  mg/kg), site B ( $37.5 \pm 24.3$  mg/kg) and residential area ( $27.9 \pm 14.5$  mg/kg). These values were higher than Pb in the cultivated soils (19.4 mg/kg) [41] and Malaysia topsoil ( $26.4 \pm 21.9$  mg/kg) [14]. The concentration also higher than the Pb in the earth crust (15 mg/kg) [40]. High Pb detected in the topsoil in this study could be related to anthropogenic activities such as atmospheric deposition from waste dumping activities and roadways [13,22,42].

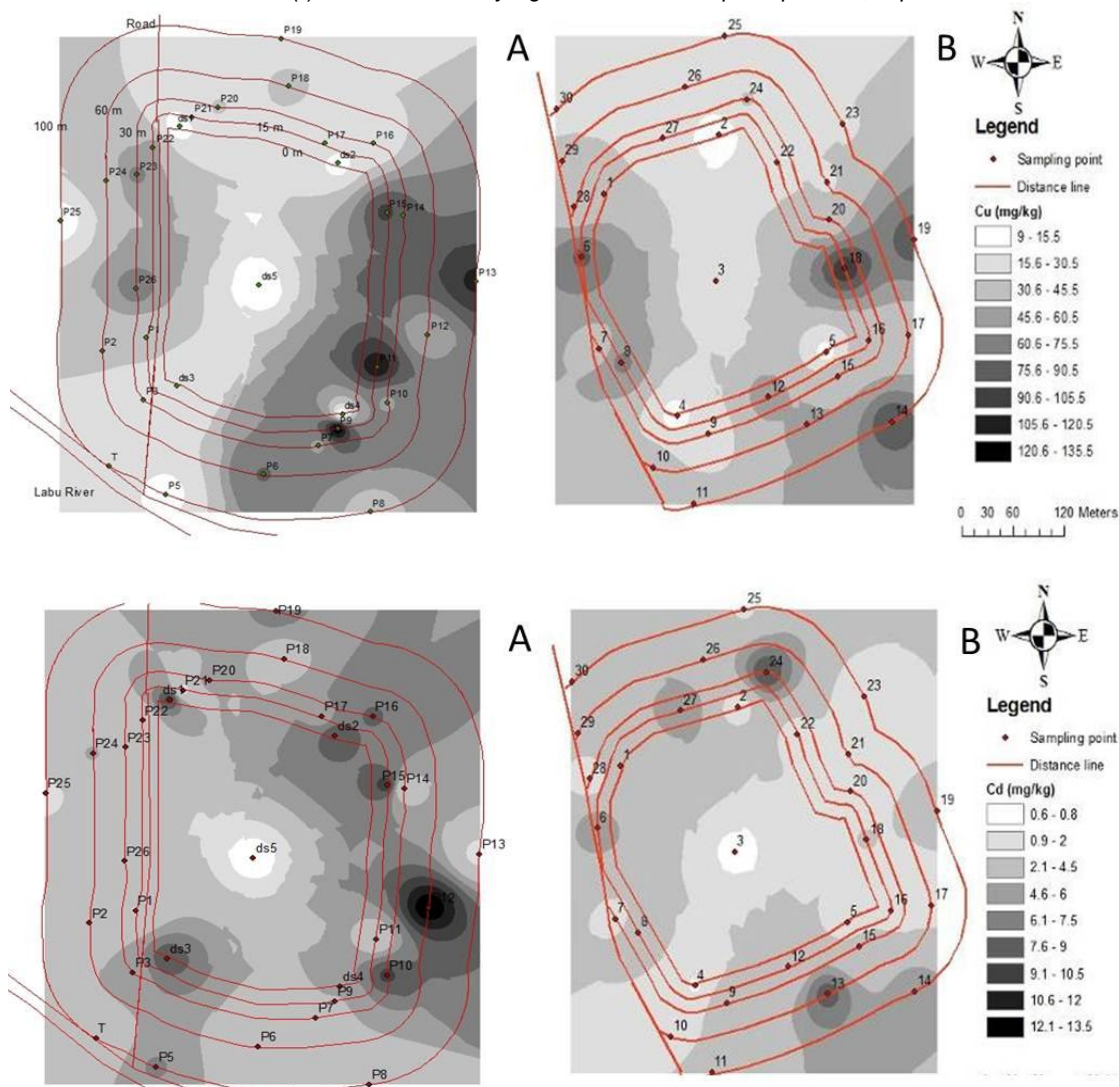
The average concentration of Zn in this study was between 4.06 and 17.8 mg/kg. It was slightly lower than the average concentration in Malaysia topsoil (2.9 to 137 mg/kg) and in uncultivated soil (18.4 mg/kg) [41]. The trend shows that Zn in the landfills was higher than other areas in this study. Zn is associated with the use of agrochemicals such as fertilisers and pesticides, and was affected by variations in soil pH and organic matter [14,39,40]. Zn distribution in soil is also control mainly by soil pH (60%) and organic matter (15%) [40].



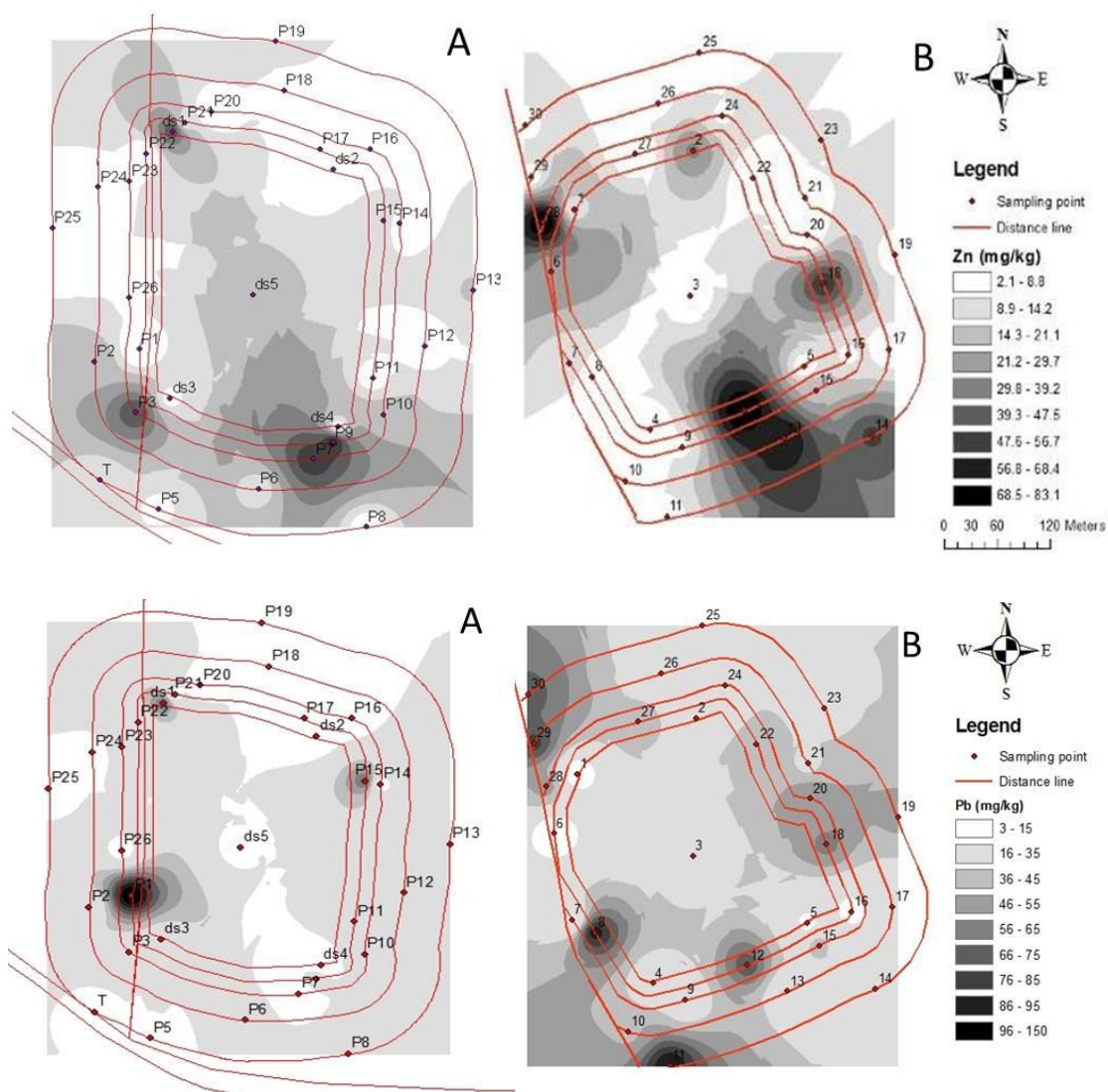
**Table 3. The correlation coefficient (r) values that indicate the associations between elements and soil properties (n = 127). Only significant relationships are shown**

	Clay	Silt	Sand	Soil pH	Organic matter	Mg <sup>+</sup>	Ca <sup>+</sup>	K <sup>+</sup>
Al	0.52**	0.47**	-0.54**	-0.76**	0.62**	0.19*		
Fe	0.38**	0.36**	-0.41**	-0.39**	0.53**	0.43**	0.26**	0.25*
Cu	0.44**	0.41**	-0.50**	-0.45**	0.59**	0.33**		
Mn				-0.27**		0.50**	0.47**	0.23*
Ni				-0.38**		0.36**	0.46**	
Cd					0.20*			
Zn					0.18*	0.26**	0.30**	
Pb						0.34**	0.22*	

Note: Asterisks (\*) denote statistically significant relationship at \* p < 0.05, \*\* p < 0.001







**Fig. 2. The spatial distribution of heavy metals in site A and B**

Cd in this study was four to five times higher than the value reported in Malaysia topsoil as reported by [14] (0.01 to 2.02 mg/kg), [9] (0.12 mg/kg) and [11] (0.01 to 0.32 mg/kg) studies. The highest Cd was detected in agriculture soils ( $4.99 \pm 1.3$  mg/kg), followed by landfill site A ( $4.61 \pm 0.6$  mg/kg) and site D ( $4.39 \pm 1.0$  mg/kg). Cd in this study was also higher than in landfill soil as reported in [12] (0.7 to 1.4 mg/kg). High Cd in landfills and agriculture soils in this study may be related to pesticide and fertilisers in the agriculture site and mixed types of waste in the landfills [24,40,42]. Disposed waste such as batteries and plastic on the landfills may produce Cd in the soil [39]. Sewage sludge application and the burning of fossil fuel also possible to

produce Cd contamination in soil [39]. This element also possibly derived from the sedimentary rocks [39]. Soil derived from igneous rocks would have Cd contents of 0.1 – 0.3 mg/kg, those on metamorphic rocks would contain 0.1 – 1.0 mg/kg Cd and those derived from sedimentary rocks 0.3 – 11 mg/kg Cd. However, in general, most soils can be expected to contain < 1 mg/kg Cd except those contaminated from discrete sources or developed on parent materials with anomalously high Cd contents such as black shales [39].

Ni in this study was generally very low, with an average value between 0.01 to 1.55 mg/kg and within the concentrations cited in the literature. It

was higher in landfills compared to the other sites. Soils throughout the world contain Ni within a broad range from 0.2 to 450 mg/kg [40]. The average total content of Ni in Malaysian topsoil is 13.7 mg/kg, and range between 0.4 to 73.5 mg/kg [14]. This is slightly higher than the value detected in [43] in a range of 0.4 to 41.3 mg/kg. High Ni was determined in paddy field (13.2 mg/kg) associated to pesticide and fertilizers application [14]. Low concentration of Ni was detected forest soils and remote areas with an average of 2.2 mg/kg [14]. However, the cultivated soils determined in [41] have slightly lower Ni with an average of 7.9 mg/kg compared to the [14] study. Ni also was detected as very low in landfill soil from Selangor, between 0.8 to 1.5 mg/kg [12]. [44] has determined the average Ni in seven landfill sites in Kuala Lumpur landfills was between 0 to 78.1 mg/kg.

Soil properties were also found to influence the concentration of heavy metals in soil. The correlation analysis in this study had suggested that acidic soils, with high organic matter and clay, contained more heavy metals compared to alkaline soil. These results were consistent with other findings such as in [13,45,46,47,48,49].

The spatial mapping highlights the distribution of Cu and Cd in the landfill boundaries, which were associated with high acidic soils. Despite some strong directional patterns in site A there were rarely simple distance decay trends, sometime due to the strong influence of individual spot values on the interpolated surfaces. Other elements i.e. Pb and Zn, also have similar distributions to Cu and Cd. High heavy metals at the boundary of landfills were possibly due to the migration of these elements from the immediate dumping area. Study by [50,51] have indicated that toxic elements can migrate up to two miles from landfill sites. Contaminants were also reported to have migrated up to one mile from a landfill in Keizer, Oregon [52]. Transportation of these contaminants can happen through sorption and migration processes, influenced by soil physicochemical properties such as pH [51]. High acidic soil at many landfill boundaries encourages heavy metals to be concentrated in this area compared to the immediate dumping ground. This is because heavy metals are more soluble in acidic soil and these elements will be likely available to plant, soil and ingested by humans [40]. Migration of contaminants is also possible in the form of leachate seepage from the waste layers [11]. Leachate contains more than 400 toxic elements which include heavy

metals [6]. Landfills in this study were surrounded by oil palm plantations where the fertilizers and pesticide were applied and this may be associated with high heavy metals in the soil. [14,41,43] have indicated that cultivated soils in Malaysia were highly concentrated with heavy metals especially Cu and Cd as a result of phosphate fertilisers application.

Most heavy metals such as Cd tend to be present at higher concentrations in the surface horizon which is partly a reflection of the inputs from atmospheric deposition, fertilisers and cycling through plants [39]. For example, studies on Mollisols and Alfisols determined the mean concentrations of 0.39 mg/kg Cd in the surface horizon and 0.23 mg/kg in the sub surface horizons. Cd and other heavy metals also were remained in the top 15 cm soil of sewage sludge amended soils. Due to this nature, together with soil chemical process, it can affects the availability of heavy metals for uptake by plants and important in consideration of the impact on human health. Relatively large concentration of Cd for instance can accumulate in edible portions without the plant showing system of stress. Cd in soils contaminated from inorganic sources such as metalliferous mining and smelting tend to be more readily accumulated in the edible portions of vegetables compared to from soils amended with sewage sludge. These elements were trace elements which are readily translocated to plant tops after absorption through roots. In addition, to uptake through the roots, Cd can effectively absorbed into foliage and translocated around plants and this is significant route for Cd into the food chain [39].

The strength of this study includes the random sampling of soil around the landfill which allow uniform distribution of sampling points – and allow preliminary statistical and geo-statistical analysis to be performed. This study extracted heavy metals in a 'mobile or ion' form with disodium EDTA reagent. The complex extracts EDTA are more attractive alternatives compared to acids or bases because they can form strong metal-ligand complexes and are thus highly effective in extracting toxic element from soils [53,54]. Measuring the 'total' content of heavy metals does not explain the mobility or availability of these elements to plants or crops, and the character of chemical bonding in soil particles [43,55,56] indicated that the severity of pollution not only depends on the 'total' content, but also the proportion of heavy metals in mobile and bioavailable forms [56] which was detected

in this study. This is because heavy metals in soils exist in the form of ions which are bound to solid phase and mobilised into the solution phase by changes of soil pH, temperature, redox potential, soil organic matter decomposition, soil texture, leaching, ion exchange processes and by microbial activity [56-59].

## 5. CONCLUSION

The measured heavy metal contamination load in the top soil is in the following order; Al > Fe > Mn > Zn > Cu > Pb > Cd > Ni. The soil contamination occurred in both landfills and non-landfill sites in this study. Cu was found to have polluted the agriculture soil, residential area and landfill site A while Cd has polluted in all study sites. This assessment has illustrated the contaminants movement horizontally towards the landfill boundary.

## COMPETING INTERESTS

Authors have declared that no competing interests exist.

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