POSSIBLE SOURCE AND PATTERN DISTRIBUTION OF HEAVY METALS CONTENT IN URBAN SOIL AT KUALA TERENGGANU TOWN CENTER

Foo Toon Fong, Poh Seng Chee, Asrul Azani Mahmood and Norhayati Mohd Tahir *

Department of Chemical Sciences, Faculty of Science and Technology
Universiti Malaysia Terengganu (UMT)
Mengabang Telipot, 21030 Kuala Terengganu, Terengganu

**Corresponding author: hayati@umt.edu.my*

Abstract

Total concentration of five trace metals (Cu, Mn, Cd, Pb and Zn) and two major elements (Al and Fe) as well as soil parameters (soil organic matter, pH and cation exchange capacity) were measured in soils of Kuala Terengganu town center. 40 surface soils (0-20cm) were collected during the month of August, 2005. The soil samples (< 600µm) were subjected to acid digestion and the concentration of total metal was measured using Atomic Absorption Spectrometer. Results show that the range of metals observed were 4.16 - 40.90 $mgkg^{-1}$, 83.70 - 380.80 $mgkg^{-1}$, 2940.00 – 28600.00 $mgkg^{-1}$, below detection limit (BDL) - 4.88 $mgkg^{-1}$, 20.00 – 219.00 $mgkg^{-1}$, 7.47 - 171.00 $mgkg^{-1}$, and 8840.00 - 62500.00 $mgkg^{-1}$ for Cu, Mn, Fe, Cd, Pb, Zn and Al, respectively. Factor and Pearson's correlation analyses suggest that the Fe, Mn and Al originates from the parent materials, whereas the possible sources of Cu, Cd, Pb and Zn are due to anthropogenic input such as vehicular traffic and metal corrosion since there are no major industrial activities in Kuala Terengganu. In addition, calculation of enrichment factors (Efs) for trace metals showed that Pb, Cd and Zn were significantly enriched, providing additional support to the contention that Pb, Cd and Zn level in Kuala Terengganu town center soils are due to human related activities.

Keywords: Surface soil, Kuala Terengganu Town Centre, trace metals, major element, AAS

Introduction

Urban soils, sometime called as "developed land", are those soils found within a city, town or metropolitan area. Urban soils have been greatly altered by construction, excavation, contamination and other activities (Plaster, 2003). Urban soils consequently are often contaminated by these human activities in urban areas; currently nearly half of the world's population lives in urban agglomeration. Human induced contamination is reflected in the high horizontal and vertical variability brought about by the anthropogenic influence on soil formation and development (Biasioli *et al*., 2005).

Urban soil acts as a sink for heavy metals and many other pollutants as shown by many research carried out in recent years (e.g. Biasioli *et al*., 2005; Lee *et al*., 2005; Surthland *et al*., 2000; Chen *et al*., 2005 and Li *et al*., 2001). These studies showed heavy metals loading from a number of different sources related to human activities, such as vehicular emissions, industrial discharges and urban development. Among these sources, vehicular emission is commonly known to be a significant and increasing source of soil pollution in urban environment. Heavy metals, also one of the atmospheric pollutants release from vehicular traffic, can accumulate in surface soil from elevated emissions and their deposition over time can lead to abnormal enrichment, then causing metals contamination of the surface soil (Wong *et al*., 2006). The presence of these elements at trace level and/or essential element at elevated concentration might cause toxic effect if exposed to human population. Therefore, the exposure of children to metals, generally accepted as the highest risk group in the urban population, who have a higher adsorption rate of heavy metals because of their active digestion system and sensitivity to haemoglobin, can greatly increase through ingestion of metal-laden soil particles via hand-to-mouth activities [Wong *et al*., 2006; Li *et al*., 2004). In addition, adults may also be exposed to threat since inhalation is the easier pathway for toxic metals to enter their body.

Owing to the profound health risks of heavy metals contamination and growing human's activities in urban environment, investigation of the source and distribution of heavy metals in urban soils is necessary in order to evaluate the risk to residents in the urban area. There has been a previous study of heavy metals in soil of Kuala Terengganu conducted by Hafiza in 2001 (Hafiza, 2001). Result in this study showed the concentration of heavy metals in soils taken from the town center contained highest level of Cu, Pb, Cd, Zn attributed to the anthropogenic input. One of the limitations of her study was that only twenty-two soil samples were collected at selected location over the Kuala Terengganu; consequently the sampling protocol might be biased. Thus, the aims of the present study are to further determine the distribution and concentration of heavy metals in soil of Kuala Terengganu town center and to evaluate the most possible sources of the heavy metals in these soils by using a appropriate sampling strategy and soil pollution mapping tool.

Experimental

The Study Area, Soil Sampling and Analysis

The study was conducted in Kuala Terengganu town center, one of the coastal towns located in Terengganu. Kuala Terengganu is the state administrative and royal capital city of the state. The area of capital is 605.28 km^2 with a population of 361, 000 people, which accounts for about 35% of the total state population. Main economic activities include retail and wholesale trade in food items, fabric and apparels, fisheries, agriculture, service industries, and tourism, particularly as a jumping-off point for the nearby Perhentian Islands and Redang Island (Kuala Terengganu, 2007).

A systematic sampling strategy was adopted to provide a sampling programme over Kuala Terengganu (Figure 1). The 40 km^2 study area was divided into 40 cells of 1 km X 1 km size quadrant. In each quadrant, 16 sub-samples were randomly collected from 0 – 20 cm layer using a polypropylene scoop. The collected sub-samples were then mixed thoroughly to give one composite sample representing one sampling cell. Consequently in this study, a total of 40 soil samples were collected and stored in clean polypropylene plastic bags and transferred back to laboratory for analysis. In the laboratory, debris, leaves, plant roots, litters and other coarse materials were first removed from each sample. After that, these soil samples were dried in lamina flow for 3 days. The dried samples were then gently grounded and passed through a stainless steel mesh of 0.6 mm in diameter. About 0.25 mg of 0.6 mm fine soil was then digested with HNO_3+HCl+HF (9+3+2 v/v) in a Teflon beaker and heated in a closed focused microwave wet digestion oven (Milistone, model Ethos Plus) at 200^0C for 30 minutes. After cooling, 9 ml of 10% boric acid were added to remove the fluoride residue. The digested solutions were filtered and made up to 50 ml with de-ionized water, and stored in polyethylene bottles. Concentration of Cu, Mn, Cd, Pb, Zn, Al and Fe were determined by atomic absorption spectrometer (FS 220A, VARIAN). Walkley and Black method was used to determine organic matter in soil (Schulte, 1995), soil pH in 0.02 M $CaCl_2$ was determined by glass electrode method (Sims *et al*., 1995) and cation exchange capacity (CEC) was determined using pH 7 ammonium acetate extraction method (Ross, 1995). All chemical used were of analytical-reagent grade and purified water of 18 $M\Omega cm^{-1}$ MilliQ Plus type was used throughout for dilutions and washing.

Result and Discussion

Soil Parameter

Table 1 shows the range, mean and median of the soil parameters analyzed for Kuala Terengganu town center soils. The mean soil pH at Kuala Terengganu town center was 4.99 and according to Marx (1999), soils with pH less than 5.10 can be classified as strongly acid. Meanwhile, the mean soil organic matter was 4.31 % while the cation exchange capacity (CEC) was 19.13 $meq100g^{-1}$. Comparison with data obtained by Hafiza (2001) clearly showed that soil pH in this study was lower but soil organic content in the soils were within similar range

Soil is an excellent adsorbent for both organic and inorganic compound and the major adsorbing surfaces in soil is soil organic matter (SOM) (Elzahabi *et al*., 2001).According to Marx (1999), sandy soil may have a low pH (acidic), where this condition occurs because sandy soils have low amounts of reserve acidity due to low CEC. According to Elzahabi (2001), the sorption of heavy metal in soil increased with increasing soil pH and conversely decreased with lower pH. However, in this study, correlation analysis showed no significant relation between these soil parameters with the concentration of heavy metals studied

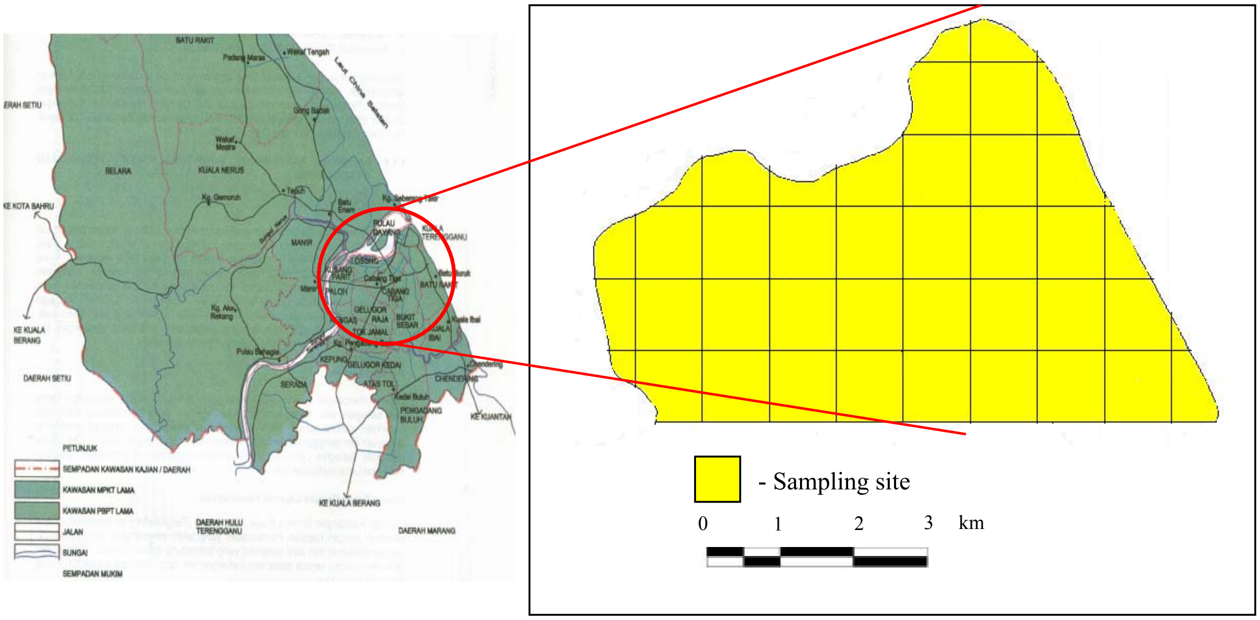

Fig. 1: Sampling area and sampling sites

Table 1: Soil parameter (Soil pH, Soil Organic Matter, and Cation Exchange Capacity) in Kuala Terengganu centre town soils and the soil parameter from previous study of Kuala Terengganu.

		Soil pH	SOM[a], %	CEC[b], meq100g^{-1}
Present study (n = 40)	Range	6.73–3.55	8.29-0.77	123 – 2.14
	Mean	4.99 ± 0.01	4.31 ± 0.19	9.92 ± 19.1
	Median	5.03	4.11	6.35
Urban soil (0 – 5cm)[c] (n = 7)	Range	6.88-4.18	9.05-1.07	-
	Mean	5.57 ± 0.93	4.38 ± 2.49	-
	Median	5.84	4.35	-
Urban soil (5 – 20cm)[d] (n = 7)	Range	7.40-4.36	7.55-1.20	-
	Mean	5.73 ± 1.09	3.09 ± 2.18	-
	Median	5.64	2.60	-

[a]Soil Organic Matter
[b]Cation Exchange Capacity
[c]Hafiza (2001), trace metal in Kuala Terengganu center town soils (0 – 5cm), Terengganu
[d]Hafiza (2001), trace metal in Kuala Terengganu center town soils (5 – 20cm), Terengganu
- Not done

Pattern Distribution of Heavy Metal Concentration

The concentrations of Cu, Mn, Fe, Cd, Pb, Zn and Al in Kuala Terengganu town center soils are summarized in Table 2. There is no information available on typical background values for Malaysian soils for heavy metal concentration in urban soils; therefore, these data were compared with available background values (means) of world average as reported by Ure and Berrow (1982), previous study of urban soils in Kuala Terengganu (Hafiza, 2001) and Seberang Perai (Lam, 2003).

The mean concentration of Cu, Mn, Fe and Al of the present study are within the lower range of background values, while Cd, Pb and Zn are located at upper range of the background values (Ure and Berrow, 1982). Compared to Hafiza's result (Hafiza, 2001), the mean concentration of Cu, Mn, Fe, Pb of the present study are lower but for Cd and Zn, the mean values obtain in this study were higher. The variation in these heavy metals concentration compared to previous study (Hafiza, 2001) could be due to the banning use of leaded petrol together with raising of land level for renovation and building construction application. All these activities may change the level of heavy metal in soil (Scharenbroch *et al*., 2005).

Comparison with the soils of Seberang Perai Tengah town center (Lam, 2003) showed that Cu, Cd and Pb in soil of Seberang Perai Tengah town center were higher than Kuala Terengganu soils. Seberang perai Tengah has relatively higher population density with a well-establish network of roads then it is not surprising that metals commonly associated with vehicular emission were higher in Seberang Perai Tengah. However, it is acknowledged higher values of heavy metals concentration were found in previous study for Kuala Terengganu and Seberang Perai could also be due to the bias of the sampling protocol in their studies where soil samples were taken only at selected location like near to roadside and the number of samples were significantly less representative than in the present study.

The soil pollution maps of the selected heavy metals (Cu, Mn, Fe, Cd, Pb, Zn and Al) were generated using SURFER 6.0. Based on these maps, 4 types of distribution pattern were identified. The first type consisted of Fe, Mn and Al (Figure 2), where the highest concentration was distributed at the agriculture site. The second type included Cu and Pb (Figure 3), where the highest concentration was distributed along the major road in the town. However, the third distribution pattern represent the distribution of Cd and Zn, respectively, which showed distinct pattern for each one as well as the first and second type of distribution map (Figure 4).

Table 2: Heavy metal concentration (mgkg^{-1}) in the Kuala Terengganu center town soils and the heavy metal concentration (mgkg^{-1}) urban soil of other city and the background values.

Location		Concentration of Heavy Metal, mgkg^{-1}						
		Cu	Mn	Fe, %	Cd	Pb	Zn	Al, %
Present study n = 40	Maximum	40.8	380	2.86	4.88	219	1700	6.25
	Minimum	4.16	83.7	0.29	BDL	20.0	74.7	0.88
	Mean	21.3	174	1.47	1.88	105	696	3.63
	Median	20.3	147	1.20	2.00	104	628	3.89
Urban soil[a] N = 7	Mean	46	984	2.23	0.78	146	65.5	-
Urban soil[b] N = 7	Mean	54.3	962	1.89	0.49	133	55	-
Urban soil[c] N = 5	Mean	118	321	0.95	3.93	118	374	1.10
Urban soil[d] N = 5	Mean	43	171	1.12	1.81	101	251	1.11
Background values[f]	Mean	25.8	761	3.20	0.62	29.2	59.8	6.65

[a]Hafiza (2001), trace metal in Kuala Terengganu center town soils (0 – 5cm), Terengganu.
[b]Hafiza (2001), trace metal in Kuala Terengganu center town soils (5 – 20cm), Terengganu.
[c]Lim (2003),trace metal in Seberang Perai Tengah center town soils (0 – 5cm), Pulau Pinang.
[d]Lim (2003),trace metal in Seberang Perai Tengah center town soils (5 – 20cm), Pulau Pinang.
[f]Ure and Berrow (1982), range of heavy metal background values (means) of soils.
BDL – Below Detection Limit.
- Data not available.

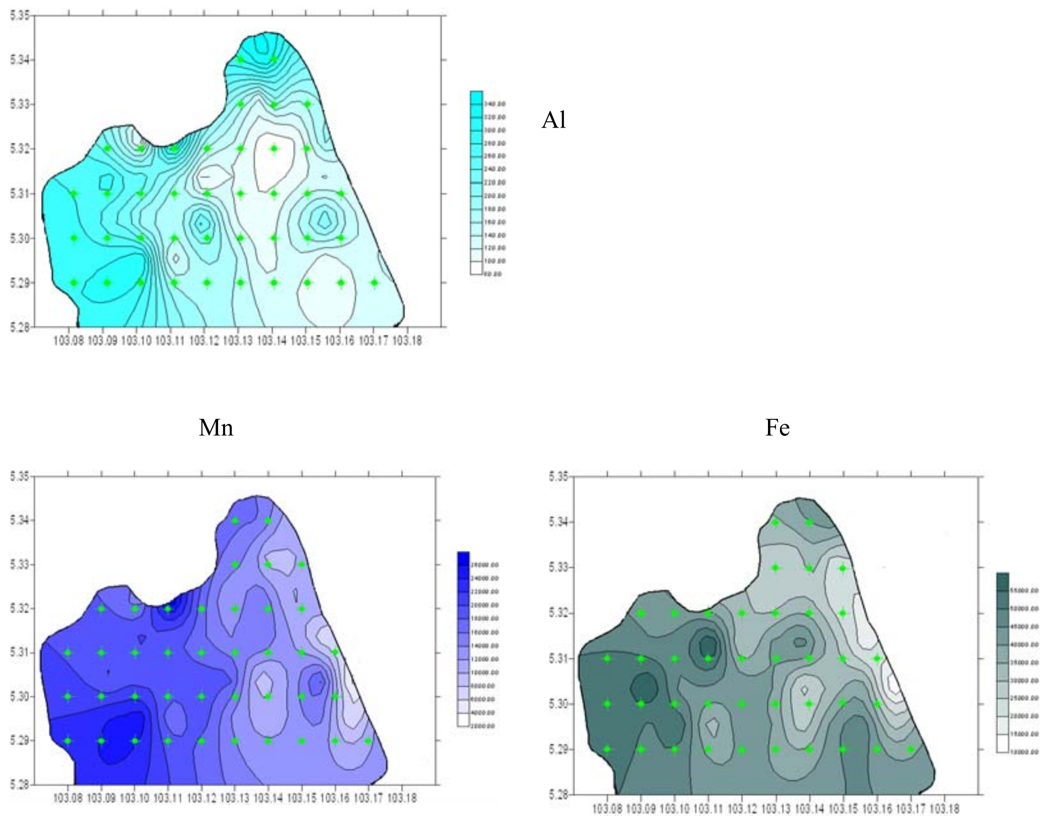

Fig. 2: The soil pollution maps of Mn, Fe and Al of Kuala Terengganu town center soils

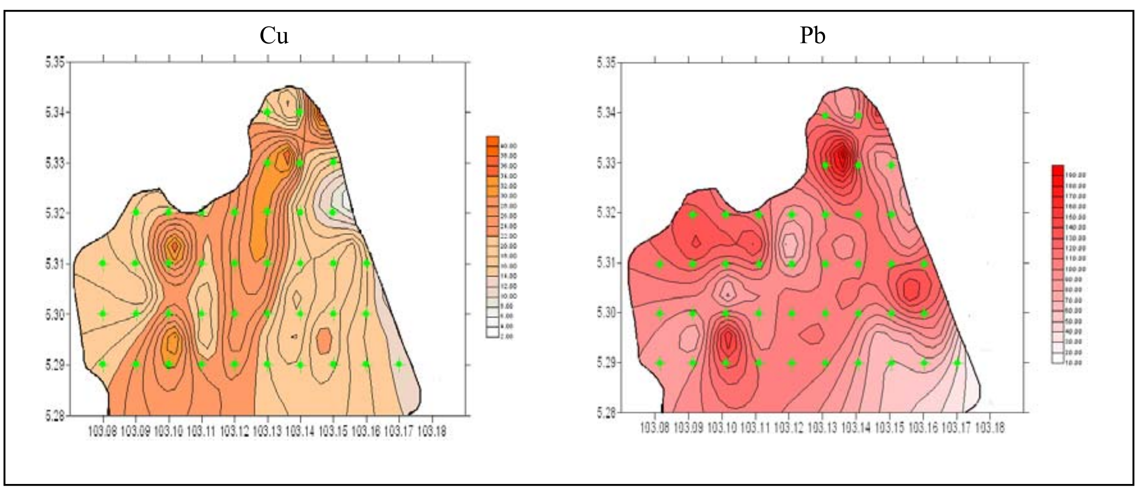

Fig. 3: The soil pollution maps of Cu and Pb of Kuala Terengganu town center soils.

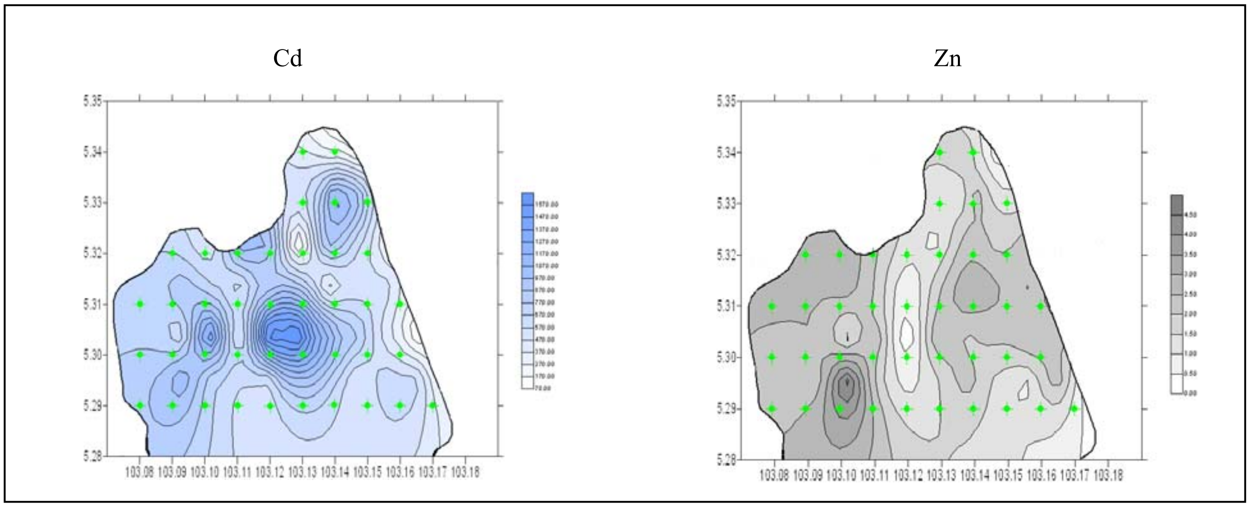

Figure 4: The soil pollution maps of Cd and Zn of Kuala Terengganu town center soils.

The Possible source of the Heavy Metals Contamination

The composition of soil at a given point is determined by contributions from various sources, such as mineralogy of the soil, deposition of combustion particles from the atmosphere, deposition of traffic-related particles, etc. The elemental composition of soil changes from one location to another, because the relative contributions of each of these sources may differ spatially (Yay, *in press*). Thus, a varimax-rotation factor analysis was applied to the data set to separate between different components of soil and the distribution of these components over the study area to evaluate the possible sources of those heavy metals in this study. Figure 3 illustrates the heavy metals concentration in the coordinate system of three factors, where three groups are obviously distinguished with eigenvalues higher than one. Elemental loadings in these factors are given in Table 3 and the factor loadings which are >0.50 are regarded as significant in the interpretation of the data. In addition, Pearson's correlation analysis was also conducted to study the relation between those heavy metals in this study. The correlation constant, r, between concentrations of the heavy metal in Kuala Terengganu center town soil is showed in Table 4.

Factor 1, which explains 35.8 % of the total system variance, has high positive factor loadings on Fe, Mn and Al and the correlation analysis showed significant correlation between Fe and Mn, r = 0.6223; Fe and Al, r = 0.6665. Fe, Mn and Al are the most abundant element in the lithosphere; Fe and Mn generally occur as Fe-Mn oxides and hydroxides which play an important role in precipitation of some heavy metals in soils while Al-hydroxides are the structure components of clay minerals. Thus, it can be concluded that Fe, Mn and Al in Kuala Terengganu town center soils are derived from parent materials (Kabata-Pendias and Pendias, 2001).

Factor 2, has moderately positive loading of Zn and a high negative loading in Cd. The negative loading of Cd reflects the possibility of anthropogenic or natural parent materials sources that contributed to the concentration of Cd in the present study. However, correlation analysis showed Cd is weakly correlated to Pb, which suggested that some fraction of the Cd could be from traffic source possibly from the deposition of particles from worn tires, because Cd is used in the vulcanization process during tire production (Schauer *et al*., 1996). In the case of Zn, it is possible that this metal is derived from mixed sources. The correlation analysis showed that Zn has weakly correlation with Fe (r = 0.3505), this suggesting that Zn could also originate from natural parent materials. However, Kuala Terengganu is a coastal town; the influence of the marine aerosol can enhance the corrosive nature of the urban atmosphere causing the corrosion of the metal (Miguel *et al*., 1997) from buildings and vehicles.

Factor 3 is also another anthropogenic contribution with high positive loading of Cu and Pb. The main source of Pb contamination in soils was derived from vehicle emission, where Pb was added to the petrol as an additive of anti-knocking in vehicle engine. Pb is a stable element in soil, and the time necessary for 10 % decrease of the total concentration by leaching was calculated to be 200 years for polluted soils and 90 years for "control" soils (Kabata-Pendias and Pendias, 2001). Thus, this might explain why even though the use of leaded petrol had been banned many years ago, the contamination in urban soils still significant due to the historical use of Pb in petrol (Lee *et al*., 2005). On the other hand, Cu contamination in urban soils could be attributed to vehicle break dust (Kouji *et al*., 2004)

Table 3: Factor analysis with rotated varimax normalized loading for measured heavy metals (loading <0.05 are not shown).

Variable	Factor 1	Factor 2	Factor 3
Cu			0.7497
Mn	0.7470		
Fe	0.9297		
Cd		-0.8401	
Pb			0.8999
Zn		0.5610	
Al	0.7914		
Eigenvalue	2.5081	1.3299	1.2052
% Variance	35.8307	18.9995	17.2182

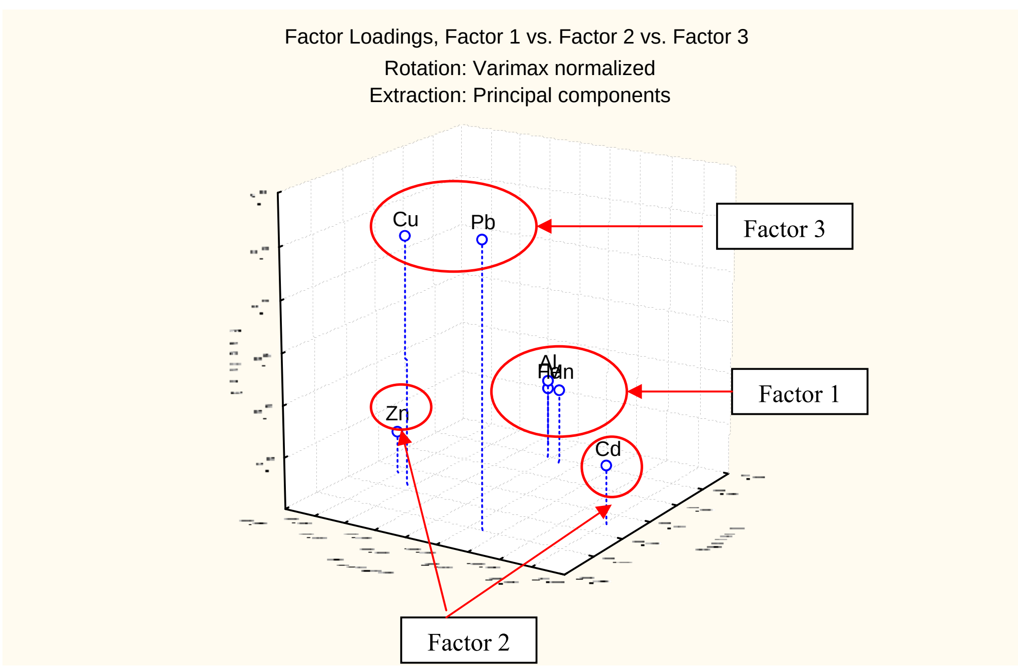

Fig. 3: Illustration of heavy metals concentrations in 40 Kuala Terengganu town center topsoil samples in the coordinate system of three factors after varimax rotation.

Table 4: The correlation constant, r, between concentrations of the heavy metal in Kuala Terengganu center town soil.

	Cu	*Mn*	*Fe*	*Cd*	*Pb*	*Zn*	*Al*
Cu	1						
Mn	0.1959	1					
Fe	0.3112	0.6223**	1				
Cd	-0.1367	0.1546	0.1621	1			
Pb	0.4546**	0.1422	0.0068	0.3430*	1		
Zn	0.2448	0.1201	0.3505*	-0.0437	0.0283	1	
Al	0.2918	0.4141*	0.6665**	0.1287	0.0365	0.1157	1

*Correlation is significant at the 0.05 level (2-tailed)
** Correlation is significant at the 0.01 level (2-tailed)

Enrichment Factor

Enrichment factor (EF) can be utilized to differentiate between the metals originating from human activities and those from natural procedure, and to assess the degree of anthropogenic influence. One such technique that has often been applied is normalization of a tested element against a reference one. Typically, the enrichment factor of a heavy metal is expressed by the following equation.

$$EF_M = ([M]_{sample}/[Mn]_{sample})/([M]_{ref}/[Mn]_{ref})$$

where EF is the enrichment factor of M, $[M]_{sample}$ and $[Mn]_{sample}$ are the concentration of the chemical element and Al in Kuala Terengganu town center soils, while $[M]_{ref}$ and $[Mn]_{ref}$ are the mean concentration (Ure and Berrow, 1982) of the chemical element and Mn, respectively. EF can give an insight into differentiating an anthropogenic source from a natural origin. Five contamination categories are recognized on the basis of the enrichment factor, where EF < 2 is deficiency to minimal enrichment; EF 2 – 5 is moderate enrichment; EF 5 – 20 is significant enrichment; EF 20 – 40 is very high enrichment and EF > 40 is extremely high enrichment (Sutherland, 2000).

Figure 4 illustrated the mean EFs increase in the order of Fe<Al<Cu<Zn<Cd<Pb. Basically, as the EF values increase, the contributions of the anthropogenic origins are also increase (Sutherland, 2000). Pb (Efs = 15.82), Cd (Efs = 13.30) and Zn (Efs = 5.49) were in the category of significant enrichment. Thus, from the calculation of enrichment factors (Efs), it showed that Pb, Cd and Zn were significantly enriched, which is providing additional support to the Pb, Cd and Zn level in Kuala Terengganu town center soils are due to human related activities. However, the remaining three heavy metals (Cu, Fe and Al) Efs values were in deficiency to minimal enrichment category.

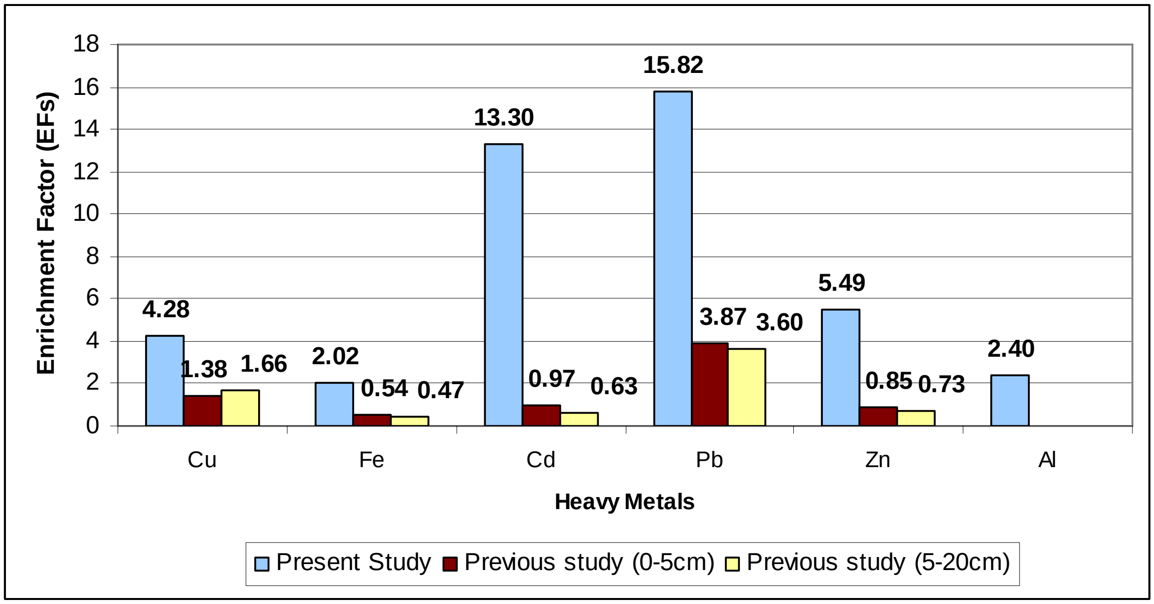

Fig. 4: Enrichment factors of selected heavy metals relatively to Al for Kuala Terengganu town center soils.

Conclusion

As a conclusion, the level of heavy metals have reduced a number in the soil of Kuala Terengganu center town, including Cu, Mn, Fe and Pb except Zn and Cd compared to previous study [9]. The concentration of heavy metals in the present study was also lower than another center town in peninsula Malaysia (Seberang Perai Tengah, Penang), which is more developed. From the research, the result prove the anthropogenic impact on Pb, Cu, Zn and Cd in Kuala Terengganu town center soils meanwhile Fe, Mn and Al were from the parent materials in Kuala Terengganu town center soils. No doubt, traffic is the major pollution source in Kuala Terengganu since there are no major industry activities in Kuala Terengganu. Thus, it is importance to further study the chemical form of the heavy metals in Kuala Terengganu Town center.

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