

Assessment of Heavy Metal Mobilization in Soils from Landfill Sites in Malaysia

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Abstract—This paper focuses on the speciation of heavy metals from two non sanitary landfills of different status in Malaysia. Leachate contaminated soils from Bukit Beruntung (BB) and Taman Beringin(TB) landfills were analyzed using sequential extraction procedure. Eight metals namely Co, Cd, Cr, Pb, Ni, Zn, Mn and Cu were considered. Analytical determinations were performed by inductively coupled plasma mass spectrometry (ICP-MS). The results show that in BB landfill, Zn, Mn and Pb were the highest concentration of metals observed in the mobile phase while only Cu and Cr concentration was predominant in the immobile phase. Co, Ni and Cd was not detected at all in BB. Similarly, in TB landfill, Mn, Zn and Pb were the highest concentration of metal observed in the mobile phase while Ni, Cu, Cr and Co were most predominant metals observed in the immobile phase. Only Cd was not detected in TB. The mobility of most metals were higher in TB compared to BB which shows that metals found in TB could pose a more serious threat to the environment. Further, this study also stressed the importance of proper investigation of ex landfill sites before any reclamation or redevelopment of the 246 ex-landfills (by the year 2020) proposed in the National Urban Policy, Malaysia.

Keywords— Heavy metals, Landfill, Mobility, Speciation.

I. INTRODUCTION

The world has been witnessing serious environmental changes especially since the dawn of industrialization. Global industrial revolution has led to the release of unprecedented amount of toxic substances (pollutants) into the environment. This has posed serious threats to human health, flora, fauna, water, soil, air and the environment as a whole (Agamuthu and Al-Abdali, 2009; Manaf *et al.*, 2009; Agamuthu and Fauziah, 2010; Duruibe *et al.*, 2007; Fong *et al.*, 2008; Kanmani and Gandhimathi, 2013; Ripin *et al.*, 2014; Mohammad *et al.*, 2011; Wuana and Okieimen, 2011; Dixit *et al.*, 2015). Natural degradation of pollutants is becoming increasingly difficult due to the quantity, complexity and heterogeneity of pollutants released into the environment. One of such ubiquitous, toxic and recalcitrant pollutant of concern is heavy metals. Examples of heavy metals includes cadmium (Cd), iron (Fe), cobalt (Co),

chromium (Cr), copper (Cu), mercury (Hg), lead (Pb), zinc (Zn), arsenic (As), manganese (Mn), nickel (Ni), selenium (Se) and the platinum group metals, which comprises platinum, palladium, rhodium, ruthenium, osmium, and iridium. There are two major sources through which heavy metals enter into the environment namely natural sources and anthropogenic or man-made sources (Mohammad *et al.*, 2011; Park *et al.*, 2011; Bolan *et al.*, 2014).

According to Ripin *et al.*, (2014) and Ashrafi *et al.*, (2014), sources and fluxes of anthropogenically generated heavy metals have received significant attention worldwide. This concern has also been raised in Malaysia. The soil environment has become major sink for these metals in most landfills in Malaysia. Although, studies on the prevalence of heavy metals in Malaysian landfills have been extensively reported (Shazili *et al.*, 2006; Bahaa-Eldin *et al.*,2008; Agamuthu and Fauziah, 2010; Emenike *et al.*, 2012;Ismail *et al.*, 2015) these studies have tended to focus on the distribution, total concentration and impact of heavy metals on surface and ground water, soils, plants, animals such as fishes and humans. The total concentration of heavy metals found in an environmental system may not necessarily indicate its toxicity, mobility and bioavailability and hence the need for chemical speciation to determine the specific physicochemical forms of heavy metals (Tack and Verloo, 1995; Nemati *et al.*, 2011). This study seeks to compare between two non sanitary landfill of different status and assess the mobility of the chemical forms of heavy metals in leachate contaminated soil from BB and TB landfill. This will provide useful information on the bioavailability, mobility and potential toxicity of these metals and will allow the assessment of the potential and actual risk posed by the non sanitary landfills.

II. STUDY AREA

TABLE I: BRIEF DESCRIPTION OF THE SAMPLED SITES

Landfill	Status	Grade	Sampled points
Bukit Beruntung (BB)	Active	Non sanitary	BSA(inside the dumping area) BSB(open area between dumping cells) BSC(entrance to the landfill)
Taman Beringin (TB)	Non active	Non sanitary	TSA(inside the dumping area) TSB(open area between waste cells) TSC(entrance to the landfill)

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III. MATERIALS AND METHOD

A. Soil Sampling and Analysis

Soils were excavated at 0-30 cm depth from areas contaminated with leachate (sampled points in Table 1) in accordance to 2014 ASTM – 1197 standard guidelines for conducting terrestrial soil-core microcosm test. Thereafter, samples were placed in clean sterile containers and taken to the laboratory where they were oven dried. Dried soil samples were stored in labelled polyethylene containers and used for subsequent analysis.

B. Sequential Extraction

In this study, a modified sequential extraction process based on Tessier *et al.*, (1979), Tsang *et al.*, (2007) and standard ISO 11466 (1995) was performed for the determination of eight heavy metals (Cr, Mn, Co, Ni, Cu, Zn, Cd and Pb) in soils samples. An initial weight of 1.0 g was used in the sequential extraction process and all sample analyses were run in triplicates. The extraction procedures employed is detailed as as shown in Table 2.

The samples were centrifuged at 3500 rpm for 8 minutes at room temperature for each extraction and the supernatants from each extraction were subjected to ICP-MS analysis. Prior to the start of the next extraction step, 10 ml deionised water was used to wash samples and then the washing solution was discarded after centrifugation. All the experiments were carried out in triplicate to reduce systematic error.

TABLE II: A FIVE-STEP SEQUENTIAL EXTRACTION PROCEDURE

Fraction	Metal Form	Extraction conditions
F1	Exchangeable	10ml 1M MgCl ₂ , pH 7.0 (Shake 2hours)
F2	Carbonate-bound	10ml 1M NaOAc, pH 5.0 (Shake 5hours)
F3	Fe-Mn oxide-bound	20ml 0.04M hydroxylamine hydrochloride in 25% acetic acid at 96°C in water bath (6 hours)
F4	Organic matter-bound	3ml 0.02M HNO ₃ and 5ml of 30% H ₂ O ₂ at 85°C in water bath 3ml 30% H ₂ O ₂ at 85°C in water bath (3 hours) 5ml 3.2M NH ₄ OAc in 20% HNO ₃ shaking at room temperature (30 min)
F5	Residual	9ml 12M HCl, 3ml 15.8M HNO ₃ , 5ml 0.5M HNO ₃ (stand at room temperature 16hours) Heat and maintain for 2 hours

IV. RESULTS AND DISCUSSION

Potential mobility and bioavailability of heavy metals at BB and TB landfill

The F1, F2 and F3 contains the mobile fraction of heavy metal in an environment while and the last two fractions (F4 and F5) are the non mobile fractions (Tessier *et al.*, 1979). Metals extracted in the F4 and F5 are generally considered neither mobile nor bioavailable because they are bound to silicates and primary minerals in uncontaminated soils and sediments (Ashraf *et al.*, 2011). A high percentage of metal in the F1, F2, F3 fractions is therefore an indication of pollution in an area.

Kabala and Singh (2001) reported that weakly bound metal fractions are the readily available fraction. Therefore, the mobility and/or immobility of heavy metals along with their availability in soil largely depends on their type of binding forms and is used as a measure of the degree of environmental pollution. High metal concentration in the mobile fraction indicates that less concentration is left in the soil (Ashraf *et al.*, 2011). The concentration of metals obtained from ICP-MS was converted into percentage as illustrated in Figure 1.

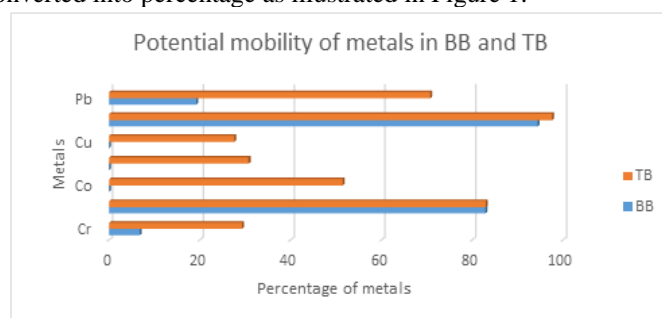


Fig. 1: Potential mobility of metals in BB and TB landfill

The trend in the mobility of the heavy metals shows that Zn, Mn and Pb have the same order of mobility in both landfill. This could be due to the similarity in the composition of waste materials received in both landfills and the non sanitary condition that prevail in both landfill allows the mobility of Zn, Mn, Pb to supersede others metals studied. Metals in TB showing more mobility than in BB and this could be due to the age and status of the landfill. The buried waste in TB (an inactive and closed landfill) has undergone a longer period of decay and thus more metal has leached out from waste material buried in the soil while BB landfill sites due to its continued activity may not have undergone the phase of acid formation when metal ion concentration is usually high. The following is a brief discussion on the mobility of each metal in both landfill.

A. Chromium

In this study, Cr in the inactive landfill (TB) was mostly found inside the dumping area and in the immobile form. This is similar in the active landfill (BB) where Cr was also found to be in the F4 and F5 fraction, however, in BB, the percentage distribution of this metal was mostly found in an open area within dumping cells. Because metals associated with the residual fraction (F5) are likely to be incorporated in aluminosilicate minerals, they are unlikely to be released to pore-waters through dissociation and therefore not readily bioavailable.

B. Manganese

The distribution patterns of Mn shows it was mostly present in the mobile form in TB and BB. High percentage of this metal in the mobile form could lead to it being bioavailable and thus enter the food chain.

C. Cobalt

In TB, Co was mostly found in the immobile form at the entrance to the landfill. Cobalt was not detected in BB landfill in all stations.

D. Nickel

Similar with Co, Ni was not detected in BB landfill but in TB it is mostly found in the mobile form in the entrance to the dumping area.

E. Copper

Copper was mostly found in the immobile form in BB and TB inside the dumping area.

F. Zinc

Zn was predominantly found in the mobile fraction in BB and TB in all sampled sites.

G. Lead

This metal was mostly found in the immobile form in BB while in TB, it was mostly present in the mobile fraction. In TB, environmental toxicity due to Pb is thus very likely due to its mobility and bioavailability

H. Cadmium

Cd was not detected at all in BB and TB landfill.

V. CONCLUSION

The chemical speciation of heavy metals in soil from BB and TB landfill revealed the mobile and non mobile forms of metals found in both landfill. Pb, Mn and Zn were the most mobile metals observed in both landfills and due to their potential bioavailability, they present an immediate risk due to the environment.

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