Multivariate analysis of the effects of age, particle size and landfill depth on heavy metals pollution content of closed and active landfill precursors

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Abstract: Multivariate analysis of a heavy metal pollution survey of closed and active landfill precursors was carried out in order to compare environmental risk levels in relation to age, particle size and depth of the precursors. Landfill precursors (77) were collected and analyzed for 15 USEPA toxic heavy metals using ICP-MS. Heavy metals concentrations in closed landfill precursors were significantly higher than those in the active landfill for 11 of 15 heavy metals investigated (closed landfill order: Fe > Al > Mn > Cu > Pb > Ba> Co > Cr > Ni > Cd > As > Se > Ti). Cluster analysis and correlation studies indicated the distribution of the metals was more influenced by landfill precursor size than by depth of the sample. Principal component analysis (PCA) showed that 10 of 15 of heavy metals of both landfill precursors were from similar anthropogenic sources. Heavy metals pollution indices (I_{geo} > 5, EF > 40 and CF > 7) of both active and closed landfill precursors exceeded limits in the order of Zn > Cd > Pb > Cu > Ag, indicating a major potential health risk influenced by age and particle size of precursor. Zn, Cd, Cu and Pb of both landfill precursors exceeded the USEPA set standard for assessment of human health risk for each of the metals (1×10^{-4} to 1×10^{-3}). This study highlights the need for the integration of a clean-up process for precursors from both types of landfill to reduce possible environmental pollution during a reuse process.

Keywords: landfill precursor, pollution index, particle size, heavy metals, human health risk, landfill

Introduction

Heavy metal deposition into landfill is of major concern due to the possible complex pathways into the environment and the possible high risk effect on living organism within the landfill areas. Heavy metal contamination from landfills has been attributed to farmland, surface water and underground water pollution (Lu et al., 2010; Chen et al., 2015, Sharifi et al., 2016).

Unlike organic pollutants, heavy metals do not degrade in the landfill and their residual time in a municipal landfill can be for about 150 years if the metal is leached at a rate of 400mm/year (EU, 2002). This indicates that only a small proportion of the possible heavy metals content of a landfill is reflected in its leachate. Major heavy metals content of the landfill is reflected by landfill precursor which is the solid waste formed as result of the heterogeneous interaction between disposed wastes, climatic conditions and the management practice of the landfill. The growing interest in landfill mining and reuse of landfill precursors as compost (Masi et al., 2014; Rong et al., 2017), landfill covering (Jain et al., 2005) and energy recovery (Quaghebeur et al., 2013) requires an evaluation of heavy metals enrichment level and associated health risks of landfill precursors, as part of a strategy to prevent further deposition of the heavy metals into the environment. Exposure to certain concentrations of heavy metal could lead to diverse health challenges especially for vulnerable people (children and aged), e.g. Cd, As, and Pb induces carcinogenesis of organs like lungs, kidney, bladder and skin (Kamunda et al., 2016).

In Nigeria, heavy metal percolation into wells and underground water within 50-100m from an active landfill at Olushosun, Lagos, had been reported (Aboyeji and Eigbokhan, 2016). The rapid urbanization in the commercial capital Lagos has also increased pressure on the government to seek alternative reuse of closed landfill precursors, but heavy metal

contamination levels and the possible human health risk involved is essential information needed to make an informed decision. Heavy metal concentrations of the landfill within the Lagos area had been largely determined by the soil/fine components of landfill, while the possible contribution of other component of the landfill has been ignored. Jain et al. (2013) and Kaartinen et al. (2013) have reported size grouping of landfill precursors as important to understanding pollution assessment and possible reuse option. Multivariate analytical tools have been deployed to measure relationship, impact and association within several symmetrical and asymmetrical environmental components (Lu et al., 2010; Singh & Kumar, 2017). There is also a paucity of published report on the effect of landfill depth and age on the heavy metal pollution indices of landfill precursors.

We report here on a multivariate analysis of heavy metals pollution survey of a closed and active landfill precursors using major pollution indicators (geo accumulation index, I_{geo}; enrichment factor, EF; contamination factor, CF), in order to compare the environmental risk levels in relationship to the age, particle size and depth of the landfill precursors.

2.0 Material and method

2.1 Sampling locations

The Olusoshun active landfill site is located in the northern part of Lagos Metropolis within the Ojota area of Ikeja Local Government Council, within a Longitude of 6 ° 35' 50"E to 6 ° 36' 30 "E and Latitude 3 ° 22 ' 45 "N to 3 ° 23 ' 30 "N. It has been in operation since November 1992 with an area of 42 hectares and receives an average of 8,000 metric tons of waste daily (Lawma, 2012). The Abule-Egba closed landfill is located in the Western part of Lagos, under

72 the Alimosho Local Government Council, with an area of about 10.2 hectares. It started receiving
73 waste in 1984 and has an estimated 1.3 million metric tons of waste with an average height of
74 12.5 m. The site had been closed since 2009 (LAWMA, 2012). Detailed site operational activities
75 of the two sites are reported in Adelopo et al. (2017). The two landfills have similar anthropogenic
76 activities around their vicinity with residential, commercial and industrial settlements bordering
77 different ends of the landfill sites. Figures 1 shows the sampling locations.

78 **** Figure 1 here ****

79 **2.2 Sampling Profile**

80 Sampling for this research was designed to evaluate the first receptor layer (between 5 and 30
81 cm) of the landfills, which reflect the early changes in the composition of the landfill waste. A
82 shallow landfill sampling covering the whole expanse of the landfill was used to reveal the
83 spatial-temporal nature of heavy metal load of waste components within this landfill layer.

84 **2.3 Sampling procedure**

85 The sites were systematically gridded into seven sampling cells using a procedure described by
86 Resource Conservation and Recovery Act (RCRA) waste sampling technical guideline
87 (USEPA, 2002). A sampling cell was approximately 14,571 m^2 for the closed landfill and
88 52,857 m^2 for the active landfill. Each cell was located using the GPS and a total of three
89 samples were obtained from each cells at different locations at the following depth: (i)
90 upperdepth between 0-15 cm; (ii) mid-depth between 16–35 cm; and (iii) low-depth between
91 36-50 cm. Sample collection was achieved using a bucket auger and samples were placed in
92 decontaminated plastic containers. An average of 500 g of sample was collected from each
93 sampling point and a total 44 samples was collected from active landfill and 33 samples from

94 the closed landfills. Oven drying, sieving and sorting were carried out in the laboratory. The
95 dried samples were separated by size into composites of less degraded (S >6.3mm) and more
96 degraded (S <6.3mm) components. A composite representative sample of 12 samples per each
97 landfill was achieved by combination of all samples of the same depth and particle size before
98 homogenization.

99 Homogenized samples (20 g) were further grinded using a mortar and pestle, and pass through
100 a uniform sieve. The powdered samples (0.5 g) were then digested by AnalaR grade acids (9.0
101 mL HNO_3 and 3.0 mL HCl) using a MARS microwave digestion system (CEM, USA) according
102 to EPA method 3052 (USEPA, 2007). Samples were filtered and diluted with distilled water to
103 the 50 ml mark, then centrifuge at 3000 rpm for 7 minutes. Aliquots of final solutions (5 ml)
104 were analysed for heavy metal content by Inductively Coupled Plasma – Mass Spectrometry
105 (ICP-MS; Agilent 7500, Agilent, USA). The instrument was calibrated prior to each set of
106 measurements. A total of 15 metals were selected based on USEPA carcinogenic potential rating
107 of metals and metalloids pollutants; Ag, Al, As, Ba, Cd, Co, Cr, Cu, Fe Mn, Ni, Pb, Se, Ti and
108 Zn were investigated.

109

110 **2.4 Quality control samples**

111 Quality control samples were digested alongside each batch of samples: spiked sample with 20
112 mg/l standard solution of Cr, duplicate sample, reagent blank and certified reference standard
113 soil sample, CRM051-50G. Microwave digest power calibration was carried out to determine
114 the optimum digestion power condition. The geochemical background value in average shale
115 was used as reference control values (Turekian and Wedepohl, 1961). The densely populated

area around the landfill had different anthropogenic activities contaminating the environment making the area unsuitable for obtaining a control sample; metal air pollution within the Lagos sampling area has previously been reported (Oketola et al., 2007).

2.5 Statistical analysis

Similarity and trends in the concentrations of heavy metals of the closed and active landfill were studied using SPSS 21. Normality test, Mann–Whitney test, cluster analysis and principal component analysis (PCA) were used to investigate the type of relationship between heavy metals concentrations of the landfill samples with the depth and size of precursors. Cluster analysis was used to sort data into groups for better understanding of the relationships between variables, and for informing further analysis.

2.6 Heavy metal pollution assessment

2.6.1 Geo-accumulation Index

The geo-accumulation index (I_{geo}) was used to estimate the metal accumulation levels in the landfill precursors. Li et al. (2014) and Aiman et al. (2016) have used this index to determine the extent of the metal accumulation in soil and environmental components above the expected natural level.

I_{geo} is expressed as:

$$Igeo = \log_2\left(\frac{Cx}{1.5Bx}\right) \quad \text{..... equ. (1)}$$

Where C_x is the concentration of the heavy metal x in the landfill precursor, and Bx is the geochemical background value in average shale of element x (Turekian and Wedepohl, 1961). The constant 1.5 is to minimize the effects of lithologic variations and small anthropogenic influences in the background values (Aiman et al., 2016; Ali et al., 2013). I_{geo} classification

according to Loska et al. (2004) and Aiman et al. (2016) is given as: unpolluted, I_{geo} <0; unpolluted to moderately polluted, $I_{geo} \leq 1$; moderately polluted, $1 \geq I_{geo} < 2$; moderately polluted to highly polluted, $2 \geq I_{geo} < 3$; highly polluted, $3 \geq I_{geo} < 4$; highly polluted to very highly polluted, $4 \geq I_{geo} < 5$; and very highly polluted, $I_{geo} > 5$.

2.6.2 Contamination Factor and Degree of Contamination

The contamination factor (CF) and the degree of contamination (DC) was use for the assessment of landfill precursor contamination. The concentrations of metals in the landfill precursors are compared to the background values of the reference sample. CF is the single metal index, while the sum of contamination factors for all metals evaluated is represented as CD. The equation for CF and CD is given by Chen et al. (2015) and Sharifi et al. (2016) as thus:

$$CF = \frac{Cs}{Cr} \quad \text{..... equ. (2)}$$

$$CD = \sum_{n=1}^{n} CF_n \quad \text{..... equ. (3)}$$

Where C_s is the concentration of each metal in the landfill precursor, while C_r is the concentration of metal in the reference control sample, as given by Turekian and Wedepohl (1961) and Ali et al. (2013). CF values are classified as low degree of contamination (CF < 1) to very high degree of contamination (CF ≥6).

2.6.3 Enrichment factor

The enrichment factors (EFs) of heavy metals were calculated to assess the contributions from anthropogenic sources to the landfill precursor concentrations. The EF is determined by

comparing the concentration of metals from the landfill precursors to that of a reference metal (Lu et al., 2010). The EF of each heavy metal in the precursor was evaluated as:

$$EF = \frac{\frac{Ci}{Cr}}{(Bi/Br)} \quad \ldots\ldots\ldots\ldots \text{ equ. (4)}$$

Where: Ci and Cr are the concentrations of the metal of interest in the landfill precursor and the chosen reference metal of the sample, respectively; Bi and Br are the background concentrations of the metal of interest in the shale and the chosen reference metals of the shale, respectively (Hu et al., 2013). The most common reference metals are Sc, Mn, Ti, Al, and Fe (Schiff and Weisberg 1999; Sutherland, 2000; Ali et al., 2013). Mn was chosen as the reference metal for the landfill precursor due to its prevalence in all samples evaluated. EF is classified as deficiency to minimal enrichment (EF ≤2), moderate enrichment ($2 \leq EF < 5$), significant enrichment ($5 \leq EF < 20$), very high enrichment ($20 \leq EF < 40$) or extremely high enrichment ($EF \geq 40$).

2.7 Potential human health risk of metals in the study sites

Heavy metals are classified as either non-carcinogenic or carcinogenic in health risk assessment (USEPA 2002a; Kamunda et al., 2016), and the potential risk procedure is calculated based on these classification. Non-carcinogenic chemicals are presumed to have threshold concentrations below which there are no potential adverse health effects, while carcinogens are assumed to have no concentrations exposure limit. The human health risk

effect of landfill precursors were assessed using the procedure provided by USEPA (1989 and 2002a) for risk exposure to heavy metals contamination on children and adults. The guidelines identify three exposure route: (a) ingestion of substrate dust particles (ADI ingestion); (b) inhalation of suspended dust particles through mouth and nose (ADI inhalation); and (c) dermal absorption of heavy metals in particles adhered to exposed skin (ADI dermal).

Average Daily intake for each pathway was calculated using equations 5-7 below:

$$\text{ADI ingestion} = \frac{\text{Cs X Ring X EF X ED X CF}}{\text{BW X AT}} \quad \text{.....equ. (5)}$$

$$ADI\ inhalation = \frac{Cs\ X\ Rhal\ XEF\ X\ ED\ X\ CF}{BW\ X\ AT\ X\ PEF} \quad \text{.....equ. (6)}$$

$$ADI\ Dermal = \frac{Cs\ X\ SA\ X\ AF\ X\ ABS\ X\ EF\ X\ ED\ X\ CF}{BW\ X\ AT} \quad \text{..... equ. (7)}$$

Where: ADI (mg kg^{-1} day^{-1}) is average daily intake (ADI) through ingestion (AIing), inhalation (ADIinh); dermal contact (ADI dermal); Cs is the concentration of the elements in the landfill precursor; EF is the exposure frequency (d/y); ED is the exposure duration (years); BW the body weight (kg); AT is the average time (days); CF is the conversion factor (1 X 10202 ^{6}); *R*inh is the ingestion (mg day^{-1}) and inhalation (m^3 day^{-1}) rate for children (1- 6 years) and above 30 years for adults; SA is the exposure surface area (cm^2/day); ABS is the skin absorption factor; PEF is the particle emission factor (m^3 kg^{-1}); and AF is the soil adherence factor (mg cm^2 h^{-1}) for both children and adults.

For this study, non-residential evaluation framework data for these parameters were used to determine ADI (USEPA, 2002a; Li et al., 2014). The daily doses estimated for each metals via the exposure pathway are divided by the reference dose (RfD, mg/kg-day) of the specific metal to yield a non-carcinogenic hazard quotient (HQ), which is aggregated together to give the overall non-carcinogens health risk index (HI, Hazard Index). Whereas, for carcinogens dose, the corresponding slope factor (SF, per mg/kg-day) for each metal is multiplied by the ADI to determine the cancer risk level per each pathway and a summation to indicate the overall risk, as indicated in equation 8-9. (USEPA, 2002a; Li et al., 2014; Chen et al., 2015).

HI (non-carcinogenic) $= \sum HQ = \sum \frac{ADI}{RfD}$ …… ……..equ. (8)

Carcinogenic risk = $\sum ADI \; X \; SF$ …… equ. (9)

3.0 Result and Discussion

3.1 Quality control samples

The recovery study of spiked sampled were within 88 to 99% for the three spiked samples used, while duplicate samples replicated at RPD< 8%. Metal concentrations for the certificated referenced sample analyzed were within the predicted interval of 80% for all elements except Ag.

3.2 Statistical analysis of trends in heavy metals concentrations

Tables 1 and 2 present heavy metal concentrations of precursors from the two landfill types with depth of sampling. The landfills had similar types of heavy metals content, but with some variation in the concentration trends of the metals. For the closed landfill the concentration order was Fe> Al > Zn >Mn>Cu>Pb>Ba>Co>Cr>Ni>Cd>As>Ag>Se>Ti, compared to the active landfill order of Fe>Al>Zn>Mn>Cu>Pb>Ba>Cr>Ni > Cd > Co >As>Ag>Se>Ti.

******Table 1 and 2 here***

The heavy metal concentrations of the landfill precursors were subjected to KolmogrovSmirov and Shapiro-Wilk normality tests to identify the appropriate SPSS analysis tools for these data. Most of the data (82%) for both landfills showed significant at the $p< 0.05$ level, which indicated that the data sets were not normally distributed. Based on these results, the non-parametric Mann–Whitney U test was used to evaluate similarity between heavy metal concentrations of samples from both landfills. Of the 15 heavy metals determined, 11 (Mn, Co, Ni, Zn, Fe, Cu, Se, As, Cd, Ba and Pb) showed a statistical significant difference ($p<0.05$) between the median concentrations of these element for active and closed landfill precursors (supplementary table), while there was no such significant statistical difference ($p>0.05$) for Ti, Al, Ag, Cr.

The box plot presented in Figure 2 compares the concentrations of each heavy metal in the active and closed landfill samples.

** **Figure 2 ****

249 The box plots (Figure 2) indicated higher concentrations of heavy metals in precursors from the
250 closed landfill compared to the active landfill for all metals investigated except Cr. Heavy
251 metals availability in landfills has been associated with the nature of waste disposed, landfill
252 management practice and degradations activities (EU, 2002). The composition
253 characterization studies of these landfills precursors, reported by Adelopo et al. (2017), have
254 shown no statistical significant difference ($p>0.05$) in the composition between the active and
255 closed landfill precursors, but a comparatively high level of degradation in the closed landfill
256 which may cause elevated concentrations of heavy metals in the samples. In older landfills,
257 there is the possibility of heavy metal diffusion into the micro pore of soil and solid matter
258 through the process of (co)precipitation and (co)flocculation, and cavity entrapment (USEPA,
259 2007a). Waste degradation may reduce the weight of landfilled waste but heavy metal
260 concentration is not often reduced. Rather the metals are being redistributed by the leaching
261 process within the depth of the landfill (EU, 2002). Tye et al. (2003) and Hamon et al. (1998)
262 suggested that the aging processes could reduce the bioavailability of metals in the soil
263 component due a stronger bonding system formed within this component.
264
265 The heavy metal content of the investigated landfill precursors were compared to previous
266 published report of heavy metals of other mined municipal solid waste landfills. Except for Cu
267 and Zn, the concentrations of heavy metals in the landfill were generally lower compared to
268 reported values of mined landfill studies in other countries: Belgium (Quaghebeur et al., 2013)
269 and United Kingdom (Gutiérrez-Gutiérrez et al., 2015). This suggests that Cu and Zn are high
270 in the content of waste disposed into the Lagos area landfills, or are influenced by the existing
271 management practice which involve the use of clay soil as linking road within the landfill.

Conversely, the concentrations of metals of landfill precursor in the present study were higher compared to reported metals in other dumpsite area within Nigeria: Sagamu, Ogun state (Ogunbanjo et al., 2016); Aba, Abia state (Amadi & Nwankwoala, 2013); and Lafia, Nasarrawa state (Opaluwa et al., 2012). Similarly, heavy metals in dust from an Electronic market at Westmestar in Lagos (Adaramodu, *et al*., 2012) and power station soil in Lagos (Adeyi and Torto, 2014) had lower concentrations than the present study. This indicates that the landfill precursors in the present study have a higher heavy metal pollution potential compared to other anthropogenic sources within the country. The reason for this could be due to the fact that the landfills are the major final disposal for all types of solid waste including electronic waste (e-waste). About 80 per cent of the world's e- wastes end up in landfills across Asia and Africa (Adaramodu, *et al*., 2012). Lagos being the commercial capital of Nigeria receives over 600,000 tons of unserviceable e-waste (computers and laptops) per year imported from developed countries as donation to organizations and educational institutions but are finally disposed at the landfills (Nnorom and Osibanjo 2008).

Longe et al. (2007) reported that the landfill sites' lateritic stratification provides natural attenuation for heavy metal percolation into the ground water. This may be responsible for low percolation of heavy metals into the ground water.

3.3 Depth and size relationships of heavy metal load in precursor

The relationships between heavy metal distribution, depth of sampling and particle size were evaluated using correlations analysis (see supplementary data). There was no significant correlation between most concentrations of heavy metals in both landfill and the depth of samples. Only Cr, Ag and Ni within the closed landfill had a strong negative correlation (-

294 0.86, -0.51, and -0.71 respectively; p=0.01) with depth of sampling. This implies that Cr and Ni
295 concentrations significantly decrease as the sampling depth increases from 5 to 50 cm. For the
296 active landfill precursors, a strong positive correlation was observed for Mn and Ba at 0.59 and
297 0.62 respectively ($p< 0.05$). This may have been influenced by metal content of waste dispose
298 and bounding system of these metals within the landfill depths. Fate and transport of metals
299 within solid waste are influence by metals' complexation system with the pore water and by
300 adsorption onto molecules of the waste (EU, 2002; USEPA, 2007a). Correlation analysis
301 between heavy metal content and particle size indicated a strong negative correlation ($p<0.05$)
302 with increased particle size for Cr (-0.82), Cu (-0.77), Ag (-0.63) and Pb (-0.63) in the active
303 landfill samples and for Co (-0.63), Mn (-0.63), Pb (-0.63) in the closed landfill samples. The
304 negative correlations indicated that the concentrations for these eight heavy metals increased
305 with decrease in the particle size, within the identified landfills. This indicates the prevalence
306 of these metals at higher concentrations in the degraded samples compared to less degraded
307 components.
308
309 ** Figure 3 here **
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311
312
313 Cluster analysis:
314 For further understanding of the interaction between heavy metal concentrations, depth and
315 particle size (degraded / less degraded) a cluster analysis was carried out. Hierarchical
316 agglomerative cluster analysis was performed using Ward's method, and squared Euclidean

distances as a measure of similarity within distribution of heavy metals within each landfill samples. The analysis of the closed and active landfill samples generated similar clusters grouping (Fig. 3). The clusters consist of two main clusters having equal number of cluster member (3) for both landfill precursors. One cluster consisting of more degraded sample (2 of 3 cluster member), while the second mainly of less degraded (2 of 3 cluster members). In both landfill, the nature of precursor (more / less degraded) was found to be linearly relate to the concentrations of heavy metals rather than the depth of precursors. More degraded landfill precursors were associated with higher concentrations of metals than the less degraded precursors of both landfills. The degraded component of the landfill precursors have the potential to adsorb more heavy metals due to increased porosity within surface area and the ability to forming a stronger bonding system (EU, 2002). Quaghebeur et al. (2013) also observed elevated concentrations of Cu, Cr, Ni and Zn in fine components of mined landfill in Belgium.

3.4 Principal component analysis (PCA)

In order to identify the source trends in heavy metals of the landfill precursors (active and closed landfills), PCA was carried out using the metal concentrations as the independent variables. The Kaiser–Meyer–Olkin test values of 0.56 (for Active landfill) and 0.59 (for closed landfill) indicated that the data were above a fair level of sufficiency. Furthermore, the Bartlett's test of sphericity with an associated p value of <0.001 indicated that PCA was suitable for the data set.

The eigenvalues of the matrix was determined using varimax rotation with Kaiser

Normalization. Varimax rotation was selected to reduce factors influencing each variable for enhanced result interpretation. The PCA for active landfill precursors indicated there are four major components with eigenvalues of the factors explaining 85% of the variance in the data set, while closed landfill precursors had five factor components and eigenvalues explaining 87% of the variance. The relations among the heavy metals based on the first three principal components are illustrated in Fig. 4 and Fig. 5, while the factor loadings result, eigenvalues and communalities are presented in the supplementary data. The results indicate there are differences in the number of factor components for the precursors in the closed and active landfill. The 1st factor explains 38.2% and 35.6% of the total variance for the closed and active precursor respectively. It loads heavily on four metals (Mn, Ba, Al, Co,) for the closed landfill sample as against seven metals in the active landfill sample (Mn, Ba, Fe, Se, Zn, Ni, and As). The component in the 2nd factor for both landfill samples were mainly of four metals (closed - Ti, As, Fe, Se; active - Ti, As, Al, Cd), accounting for 15.4% and 28.8% of the total variance respectively. The 3rd factor of the landfill samples have the same variance effect of ≈12% and consist of 4 and 3 metals for closed (Cu, Cr, Ni, Zn) and active (Cu, Cr, Pb) landfill precursors. From the component grouping it could be inferred that the metal source of the 1st component of the landfill samples (active vs. closed) is the major distinguishing source between the landfill samples (7 metals vs. 4 metals), while the 2nd and 3rd components were similar in terms of numbers and types of metals in the components (4 vs. 4) and (3 vs. 4) for the closed and active landfill samples respectively. The heavy metal source in landfill samples is dependent on the component waste and mineral content of the landfill covering (EU, 2002). The sharp difference in the 1st component may be a result of the metals from the clayey landfill cover used in the active landfill which is absent in the closed landfill (LAWMA 2012;

Longe, et al. 2007).

****Figures 4 and 5 ****

3.6 Pollution indicators

3.6.1 Geo-accumulation Index

Most (10 of 15) of the heavy metals evaluated (Ti, Cr, Mn, Fe, Co, Ni, As, Se, Pb, Ba and Al) had a geo-accumulation index below zero (Igeo <0) for both closed and active landfill precursors (see supplementary data). This indicates that both landfills were uncontaminated in terms of these metals. However, the landfill precursors contained pollution concentrations levels of Zn, Cd, Ag and Cu (Igeo > 2). The Igeo accumulation of Cd for more degraded samples of both landfills indicated heavier pollution than the less degraded sample (MD =Igeo >4, LD =1 > *I*geo ≤ 2). The pollution trend bears no definitive relationship to the depth of samples in both landfill samples ($p>0.05$). Elevated geo-accumulation of Cd in landfill and dumpsite has been reported for Ogun, Nigeria (Ogunbanjo et al., 2016) and for Pakistan (Aiman et al., 2016). The level of geo-accumulation Cd metals in the landfill samples present a major health risk. Exposure to high concentrations of Cd could damage the reproductive system, lungs, DNA and kidney, and could cause deficit in learning, cognition, behaviour and neuromotor skills in children (Chen *et al.*, 2015). Disposal of e-waste, batteries and painting residual are the likely source of Cd on unregulated municipal landfill (EU, 2002).

3.6.2 Contamination factor (CF) and the degree of contamination

CF values of most metals (Ti, Cr, Mn, Fe, Co, Ni, As, Se, Ba and Al) were within the low degree of contamination category for all precursors evaluated (CF< 1) (see supplementary data). A

serious source of concern is the CF values for Zn, Cd, Cu, Pb and Ag, which fall within a very high degree of contamination (CF≥6). There was no definite depth relation with the CF, except for Ag which increased down the depth of the active landfill for both MD and LD (MD: 7- 71, LD: 4- 17). CD values for samples in both landfill indicated a very high degree of contamination (CD >28), which is mainly influenced by the CF values of Zn, Cd, Cu, Pb and Ag. For both landfill precursors, the degree of contamination of Zn, Cd, Cu, Pb and Ag in the more degraded samples was higher than in the less degraded sample.

3.6.2 Enrichment factor (EF)

The metal EFs for Ti, Cr, Mn, Fe, Co, Ni, As, Se, Al and Ba were below enrichment level (EF≤ 2) for precursors of both landfills. However, extreme high enrichment was observed for Zn, Cd, Cu Pb and Ag for both landfill sample (EF > 40). EF values of <2 are assumed to be an indication that metal are mostly from natural source (Zhang and Liu, 2002), while EF values >2 suggest a significant contribution from an anthropogenic source. The high EF values for Zn, Cd, Cu, Pb and Ag strongly indicated that the metals are from anthropogenic source, mostly from waste disposed on the landfill.

The pollution index (I_{geo} > 5, EF > 40 and CF> 7) indicated a similar trend for metals likely to pose major challenge in the reuse options of both landfill precursors, i.e. Zn, Cu, Cd, Pb and Ag. The pollution indexes imply that the landfills' (closed and active) precursors have high concentrations of heavy metal with potential human health risk. It is evident from the results that both less degraded and more degraded pose potential danger to human health.

There is an observed similarity in the trend of heavy metal pollution in the present study and previously reported trend in the contamination of metals in soils of e-waste recycling area in China (Chen *et al*., 2010; Tang *et al*., 2010; Luo *et al*., 2011). In all of these studies, Zn, Cu, Cd, and Pb are the major contaminants, although their concentrations varied depending on

411 sampling area. This could suggest the possible accumulation of e-waste at the investigated
412 landfills.
413
414
415 **3.7 Human health risk assessment**
416 Human health risk assessment of the landfill precursors focused on the possible main routes of
417 heavy metal contamination through operations on the landfill and during reuse. Three exposure
418 pathways were identified: (i) ingestion of the metals through water consumption after
419 contamination of underground / surface water or oral ingestion along with food due to poor
420 hygiene; (ii) inhalation of suspended dust particles through mouth and nose (CDIinh) during
421 clearing, spreading, sorting and excavation of waste on the landfill; and (iii) skin absorption
422 through bathing with contaminated water.
423 Five heavy metals (Zn, Cd, Ag, Pb and Cu) identified as having a high pollution index were
424 evaluated. There are no carcinogenic slope factors for Pb, Cu, Ag and Zn; only the carcinogenic
425 risks for Cd was estimated using carcinogenic slope factors, while others were compared based
426 on cumulative intakes of the metals. Table 3 and 4 present the risk factor for carcinogenic and
427 non-carcinogenic metals of landfill precursors. The result indicated that for precursors from
428 both types of landfill the potential risk was in the order dermal > ingestion > inhalation, for both
429 non-carcinogenic and carcinogenic risk factor.
430 ******Tables 3 and 4*****
431 The inhalation risk factor falls below the acceptable range of the carcinogenic risk (1×10^{-6} to
432 1×10^{-5}). However, ingestion and dermal risk factor were mostly above acceptable level (1×10^{-4} to 1×10^{-3}). According to USEPA guidelines, cancer risk factor is expressed as a unitless
434 probability with a threshold exceeding 1×10^{-5} (USEPA, 1989). Inhalation risk factor is

435 influenced by the aerodynamic size, lifetime in air and behavior of associated composition of
436 metals in the respiratory system of human (EU, 2002). Kamunda et al. (2016) reported similar
437 observations with limited risk contribution via inhalation for heavy metal within a mining site
438 in South Africa. The total cancer risk factor from all pathway indicated that Zn and Cu were the
439 most potent health risk hazards. The hazard quotients (HQs) of Pb, Cu, Zn and Ag were within
440 the acceptable limit (HQ< 1) for all precursors. Cd is potentially a major risk especially among
441 the children with HQ above 1 (close landfill precursor, 1≥HQ≤5; active landfill precursor,
442 1≥HQ≤2). The MD valued showed higher risk than the LD samples for all metals considered.
443 The risk assessment of both landfill precursors also showed similar potential risk with 90% of
444 samples from both landfill below the acceptable limit. The risk potential of the metals in both
445 landfill precursors were in order Cd> Pb > Cu > Zn >Ag.
446
447 The research reported on here provides important information that needs to be considered in
448 development of a policy for environmentally sustainable reuse of landfilled waste. The pollutant
449 indexes (Igeo, > 5, EF> 40 and CF> 7) of five metals are identified as possible source of
450 contamination. Extractive and recycling processes of landfill waste should be carried out with
451 adequate attention to preventive protective materials for the site workers and the environment.
452 Presently, most recycling activities on both landfill sites are carried out with inadequate
453 attention to potential health risk. Tang et al. (2015) reported considerable contamination of the
454 soil and sediment by Cd through recycling of plastic due to different mechanical recycling
455 processes. Waste spreading and compacting activities could increase aerodynamic size and
456 lifetime period of heavy metals in air. Landfill mining should preferably be carried out during
457 low wind speed periods to limit particulate dispersal, which could affect the air quality. The

high concentrations of these metals precludes consideration of these landfill precursor for use as compost for farmland and agricultural purposes. The more degraded (MD) and less degraded (LD) components have pollution index values that indicate a clean-up process is required before possible reuse options are considered. Singh and Lee (2015) reported that an extraction clean–up process of an automobile shredder residues waste was able to reduce high risk factor of Pb, Zn Cu and Cd to tolerant level.

The degree of clean-up needed may be under estimated if heavy metals pollution toxicity was estimated based on the more degraded part of landfill waste alone, as the data reported on here indicates that the less degraded component contributed significant pollution to the landfill.

4.0 Conclusion

Heavy metal content of landfill waste is a major challenge in developing a sustainable reuse process for landfilled waste. The heavy metals concentrations in closed landfill precursors were significantly higher than those from active landfill for 11 of 15 heavy metals investigated. The differences were likely due to the age difference between the landfill precursor (closed: 7-8 years, active: 1-2years). Though both landfill had similar heavy metals content Fe > Al > Mn > Cu > Pb > Ba> Co > Cr > Ni > Cd > As > Se > Ti, cluster analysis and correlation studies indicated the distribution of the heavy metals were influenced by precursor size (more degraded vs. less degraded) than by depth of the sample. PCA analysis indicated similar source for 10 of 15 of heavy metals investigated for both landfill precursors. The heavy metals pollution index (Igeo, EF, CF) of active and closed landfills indicated a major health risk potential, in the order of Cd > Cu > Pb > Zn > Ag.

The study showed that landfill precursor of both landfill posed a major human health risk with carcinogenic and non-carcinogenic risk of Zn, Cd, Cu and Pb above the USEPA set standard

for each of the metals. In light of previous studies (Chen *et al*., 2010; Tang *et al*., 2010; Luo *et al*., 2011), it is possible that e-wastes may be the main source of these elements. It also identified particle size as an essential factor in evaluating the potential pollution risk factor of landfills precursor for reuse. Integration of a clean-up process for the landfill precursor during any reuse process is highly recommended in order to reduce possible environmental pollution.

REFERENCE

Aboyeji O. S., Eigbokhan S.F., 2016. Evaluations of groundwater contamination by leachates around Olusosun open dumpsite in Lagos metropolis, southwest Nigeria. Journal of Environmental Management. 183, 333-34.

Adaramodu, A. A., Osuntogun, A. O., Ehi-Eromosele, C. O., 2012. Heavy Metal Concentration of Surface Dust Present in E-Waste Components: The Westminister Electronic Market, Lagos Case Study. Resources and Environment 2(2): 9-13.

Adelopo, A.O., Haris, P.I., Alo, B., Huddersman, K. Jenkins, R.O., 2017. Comparative Characterization of Closed and Active Landfill Composites Using EDX, FTIR and Proximate Techniques. Waste and Biomass Valorization, 8(4), 1313-1323.

Adeyi, A.A. Torto N., 2014. Profiling heavy metal distribution and contamination in soil of old power generation station in Lagos, Nigeria American Journal of Science and Technology. 1(1), 1-10.

Aiman, U., Mahmood, A., Waheed, Sidra, Malik, R.N., 2016. Enrichment, geo-accumulation and risk surveillance of toxic metals for different environmental compartments from Mehmood Booti dumping site, Lahore city, Pakistan, Chemosphere 144 ,2229–2237.

Ali, Z., Malik, R. N., Qadir, A., 2013. Heavy metals distribution and risk assessment in soils affected by tannery effluents. Chem.Ecol. 29(8), 676–692.

Amadi, A. N., Nwankwoala, H. O., 2013. Evaluation of Heavy Metal in Soils from Enyimba Dumpsite in Aba, Southeastern Nigeria Using Contamination Factor and Geo-Accumulation Index. Energy and Environment Research. 3,125-134.

Chen, L., Yu, C., Shen, C., Zhang, C., Liu, L., Shen, K., Tang, X., Chen, Y., 2010. Study on adverse impact of e-waste disassembly in East China by chemical analysis and bioassays. *J. Soil. Sediment.* 10: 359-367

514 Chen, H., Teng, Y., Lu S., Wang Y., Wang J., 2015. Contamination features and health risk of
515 soil heavy metals in China. Science of the Total Environment 512(513), 143–153.
516
517
518 EU, 2002. Heavy Metals in Waste final report, European Commission DG ENV. E3 available
519 online: https: ec.europa.eu/environment/waste/studies/pdf/heavy_metals reports pdfs.
520 (Accessed October, 2017).
521
522 Gutiérrez-Gutiérrez, S. C., Coulon, F., Jiang Y., Wagland S., 2015. Rare earth elements and
523 critical metal content of extracted landfilled material and potential recovery opportunities.
524 Waste Management. 42,128–136.
525
526 Hamon, R, McLaughlin, M.J., Naidu, R., Correll, R., 1998. Long-term changes in cadmium
527 bioavailability in soil. Environ Sci Technol 32(23), 3699-3703.
528
529 Hu Y., Liu, X., Bai, J., Shih, K., Zeng, E. Y., Cheng H., 2013. Assessing heavy metal pollution
530 in the surface soils of a region that had undergone three decades of intense industrialization and
531 urbanization. Environ Sci Pollut Res. 20, 6150–6159.
532
533 Jain, P., Kim, H. and Townsend, T.G., 2005. Heavy metal content in soil reclaimed from a
534 municipal solid waste landfill *Waste Management*. 25, 25–35.
535
536 Jain, P., Townsend, T.G., Johnson, P., 2013. Case study of landfill reclamation at a Florida
537 landfill site. Waste Management. 33, 109–116.
538
539
540 Kaartinen, T., Sormunen, K. and Rintala, J., 2013. Case study on sampling, processing and
541 characterization of landfilled municipal solid waste in the view of landfill mining. *Journal of*
542 *Cleaner Production*. 55, 56–66.
543
544 Kamunda C., Mathuthu M., Madhuku M., 2016. Health Risk Assessment of Heavy Metals in
545 Soils from Witwatersrand Gold Mining Basin, South Africa. Int. J. Environ. Res. Public
546 Health.13, 663- 674
547
548 Lagos Waste Management Authority (LAWMA), (2012), MSW PROJET OPPORTUNITY,
549 Lagos State, Nigeria. http://www. lawma.gov.ng/lawma_landfill.html (accessed October, 2017)
550
551 Li, Z.Y., Ma, Z.W., Kuijp, T.J., Yuan, Z.W., Huang, L., 2014. A review of soil heavy metal
552 pollution from mines in China: pollution and health risk assessment. Sci. Total Environ. 468–
553 469, 843–853.
554

555 Longe, E.O., Enekwechi, L.O., 2007. Investigation on potential groundwater impacts and
556 influence of local hydrogeology on natural attenuation of leachate at a municipal landfill. Int.
557 J. Environ. Sci. Technol. 4, 133–140.
558
559 Loska, K., Wiechula, D., Korus, I., 2004. Metal contamination of farming soils affected by
560 industry. Environ. Int. 30, 159–165.
561
562 Lu, X., Wang, Lijun, Li, L.Y., Lei K., Huang, L., Kang D., 2010. Multivariate statistical
563 analysis of heavy metals in street dust of Baoji, NW China. Journal of Hazardous Materials.
564 173, 744–749.
565
566 Luo, C. L., Liu, C. P., Wang, Y., Liu, X., Li, F. B., Zhang, G., Li, X. D. 2011. Heavy metal
567 contamination in soils and vegetables near an e-waste processing site, south China. *J. Hazard.*
568 *Mater.* 186: 481- 490.
569
570
571 Masi S., Caniani, D., Grieco, E., Lioi, D.S., Mancini, I.M., 2014. Assessment of the possible
572 reuse of MSW coming from landfill mining of old open dumpsites. Waste Management. 34,
573 702–710.
574
575 Ogunbanjo O., Onawumi, O., Gbadamosi, M., Ogunlana A., Anselm, O., 2016. Chemical
576 speciation of some heavy metals and human health risk assessment in soil around two municipal
577 dumpsites in Sagamu, Ogun state, Nigeria. Chemical Speciation & Bioavailability. 28 (1-
578 4),142-151.
579
580 Oketola, A.A., Osibanjo O., 2007. Estimating sectoral air pollution load in Lagos by industrial
581 pollution system (IPPS). Science of the Total Environment. 377, 125 – 141.
582
583 Nnorom, I. C., Osibanjo O., 2008. Electronic waste (e-waste): Material flows and management
584 practices in Nigeria. Waste Management 28: 1472–1479
585
586 Tang, X., Shen, C., Shi, D., Cheema, S. A., Khan, M. I., Zhang, C., Chen, Y. 2010. Heavy metal
587 and persistent organic compound contamination in soil from Wenling: an emerging ewaste
588 recycling city in Taizhou area, China. *J. Hazard. Mater.* 173: 653 -660
589
590 Tang Z., Zhang, L., Huang Q., Yang, Y., Nie, Z., Cheng, J., Yang J., Wang Y., Chai M., 2015.
591 Contamination and risk of heavy metals in soils and sediments from a typical plastic waste
592 recycling area in North China. Ecotoxicology and Environmental Safety. 122, 343– 351.
593

Quaghebeur, M., Laenen, B., Geysen, D., Nielsen, P., Pontikes, Y., et al., 2013. Characterization of landfilled materials: screening of the enhanced landfill mining potential. J. Clean. Prod. 55, 72–83.

Sharifi Z., Hossaini, S. M.T., Renella, G., 2016. Risk assessment for sediment and stream water polluted by heavy metals released by a municipal solid waste composting plant. Journal of Geochemical Exploration 169, 202–210.

Schiff, K.C., Weisberg, S.B., 1999. Iron as a reference elements for determining trace metal enrichment in Southern California coastal shelf sediments. Mar. Environ.Res. 48, 161– 76.

Singh, U.K., Kumar, B., 2017. Pathways of heavy metals contamination and associated human health risk in Ajay River basin, India. Chemosphere. 174, 183-199.

Singh, J., Lee, B. (2015). Reduction of environmental availability and ecological risk of heavy metals in automobile shredder residues. Ecological Engineering 81,76–81

Sutherland, R.A., 2000. Bed sediment-associated trace metals in an urban stream, Oahu, Hawaii. Environ. Geol. 39,611– 27.

Turekian, K.K., Wedepohl, K.H., 1961. Distribution of the elements in some major units of the earth's crust. Geol.Soc.Am. Bull.72 (2), 175–192.

Tye, A.M., Young, S.D., Crout, N.M.J., Zhang, H., Preston, S., Barbosa-Jefferson, V.L., Davison, W., McGrath, S.P., Paton, G.I., Kilham, K., Resende, L., 2003. Predicting the activity of Cd^{2+} andZn^{2+} in soil pore water from the radio-labile metal fraction. Geochim Cosmochim Acta. 67(3), 375-385

Rong, L., Zhang, C., Jin, D., Dai, Z., 2017. Assessment of the potential utilization of municipal solid waste from a closed irregular landfill. Journal of Cleaner Production. 142, 413-419.

USEPA 2007. Protocol 3051A Microwave Assisted Acid Digestion of Sediments, Sludges, Soils, and Oils.

USEPA 2007a. Framework for Metals Risk Assessment, Office of the Science Advisor Risk, EPA 120/R-07/001.www.epa.gov/risk/framework-metals-risk-assessment (accessed November, 2017)

USEPA 2002. *RCRA Waste Sampling Draft Technical Guidance, Planning, Implementation and Assessment*, EPA530-D-02-002, Office of Solid Waste, Washington, DC.

USEPA, 1989. Risk Assessment Guidance for Superfund. Human Health Evaluation Manual, (Part A) [R] vol. 1, EPA/540/1-89/002. Office of Emergency and Remedial Response, Washington, DC

USEPA 2002a. Supplemental Guidance for Developing Soil Screening Levels for Superfund Sites. OSWER 9355.4-24 .Office of emergency and remedial response. Washington, DC.

Figure 1: Map of the sampling Area

Figure 2: Comparison of heavy metals concentrations in the closed and active landfills.

Figure 3: **Dendrograms showing similarity in the active and closed landfill samples based on heavy metals composition.**

More degraded samples (MD); less degraded sample (LD); depth of sampling (lower, mid, upper)

Figure 4: Rotation component of heavy metals content in the active landfill precursor

Figure 5: Rotation component of heavy metals content in the closed landfill precursor

Table

Table 1 HEAVY METALS CONCENTRATIONS OF CLOSED LANDFILL PRECURSORS AT EACH DEPTH (mg/kg)

Element	Ag	Al	As	Ba	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Ti	Se	Zn
More degraded samples															
Upper layer															
Mean(N=4)	0.79	3370.08	2.81	117.77	4.48	53.31	26.15	538.84	11910.47	363.37	11.59	253.67	0.03	0.4	4316.81
SD	0.3	1288.51	0.02	87.29	1.1	30.22	6.35	344.33	608.91	66.95	3.05	46.31	0.01	0.2	1593.67
mid layer															
Mean(4)	0.71	4241.25	3.05	118.11	4.58	41.33	32.6	262.17	17488.41	373.44	14.2	184.4	0.03	0.34	3734.83
SD	0.22	1596.8	0.64	52.46	1.13	47.63	11.54	25.87	3992.65	154.95	5.87	18.63	0.01	0.07	2843.2
lower layer															
Mean(4)	1.24	4296.29	4.41	287.56	12.49	8.97	46.63	1250.15	15095.17	472.55	22.74	339.1	0.04	0.36	2443.35
SD	0.21	258.79	1.35	9.29	10.36	3.92	4.93	1334.4	2739.57	113.58	10.78	139.31	0.01	0.03	724.66
Less degraded samples															
upper layer															
Mean(4)	0.6	3245.77	2.71	82.78	3.09	15.4	18.14	170.5	16707.67	236.94	38.07	107.26	0.03	0.46	1221.95
SD	0.42	2259.44	0.98	15.2	3.33	18.39	6.45	9.13	1127.96	124.61	39.88	55.98	0.01	0.38	1198.94
mid layer															
Mean(4)	0.3	2255.58	4.21	96.11	2.66	12.16	17.09	177.68	9558.39	607.76	10.77	88.05	0.03	0.25	2126.14
SD	0.12	888.64	2.98	53.21	0.16	3.48	3.26	160.35	940.33	562.51	4.02	31.8	0	0.04	569.19
lower layer															
Mean(4)	0.39	2796.48	3.73	233.53	5.63	34.08	17.89	141.22	34130.09	7616.55	24.07	198.24	0.03	0.91	13204.81
SD	0.3	132.75	2.68	152.2	2.71	9.84	0.88	64.47	18730.28	9650.75	25.06	175.3	0	0.9	11609.91

SD: standard deviation, N: number of sample

Table 2 HEAVY METALS CONCENTRATIONS OF ACTIVE LANDFILL PRECURSORS AT EACH DEPTH (mg/kg)

Ni	Pb	Ag	Al	As	Ba	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Ti	Se	Zn
Upper layer																
Mean(4)		0.55	2930.86	2.13	68.25	1.83	3.26	29.18	168.88	15091.74	295.50	16.63	83.44	0.03	0.20	1883.31
SD		0.20	573.17	0.04	1.20	0.26	0.96	3.54	171.97	3013.10	89.10	10.78	2.71	0.01	0.02	1734.97
mid layer																
Mean(4)		0.41	2628.61	1.55	54.53	5.16	3.12	30.94	58.30	9644.72	247.86	10.50	105.39	0.02	0.19	663.23
SD		0.04	836.39	0.00	23.14	4.21	1.43	12.56	0.27	258.45	144.63	2.14	41.75	0.01	0.02	321.12
lower layer																
Mean(4)		5.00	2792.73	2.06	73.95	2.97	3.17	21.15	77.94	11184.90	183.84	10.33	82.69	0.03	0.22	965.83
SD		6.53	99.33	0.24	6.73	2.02	0.31	4.13	25.15	38.74	2.96	1.35	11.27	0.00	0.01	256.15
Less degraded samples upper layer																
Mean(4)		0.30	2434.01	1.85	48.65	0.88	1.91	35.63	42.70	10249.65	161.30	7.05	42.41	0.02	0.20	372.89
SD		0.20	1474.46	1.18	24.18	0.65	0.54	13.60	38.63	5401.86	102.41	6.33	11.28	0.01	0.04	268.37
mid layer																
Mean(4)		0.40	3681.89	2.33	59.52	2.96	2.81	22.66	76.87	11263.09	245.59	9.15	93.67	0.03	0.22	563.86
SD		0.16	1707.35	0.42	2.39	1.41	0.47	6.10	41.32	4233.93	84.63	3.60	37.72	0.00	0.07	122.51
lower layer																
Mean(4)		1.22	1828.37	1.55	48.34	2.35	1.37	13.11	97.41	10511.49	105.91	5.34	40.51	0.03	0.17	1454.36
SD		1.38	161.52	0.47	12.39	2.05	0.23	1.78	82.89	6458.32	21.25	1.57	5.22	0.01	0.01	1343.69

SD: standard deviation, N: number of sample

Table 3 POTENTIAL HUMAN HEALTH RISK ASSESSMENT INDEX OF HEAVY METALS OF CLOSED LANDFILL

PRECURSORS (Non-carcinogenic and carcinogenic)

Age group	Element	Cu			Cd			Pb			Ag			Zn		
Adult																
More degraded samples																
		mean	mini	max	mean	mini	max	mean	mini	max	mean	mini	max	mean	mini	max
	ingestion	3E-02	1E-02	5E-02	1E-02	5E-02	2E+00	1E-01	8E-02	2E-01	3E-04	2E-04	4E-04	2E-02	1E-01	2E-02
HQnc	inhalation	4E-04	3E-05	9E-04	2E-05	9E-06	3E-05	2E-04	1E-04	2E-04	4E-07	1E-06	2E-06	2E-05	2E-05	3E-05
	dermal	3E-02	1E-02	6E-02	4E-01	3E-01	7E-01	3E-02	2E-02	4E-02	5E-07	4E-07	7E-07	3E-02	2E-02	4E-02
	HI_C	8E-04	6E-14	6E-04	8E-06	2E-14	6E-06	4E-05	8E-15	4E-05	8E-07	5E-16	9E-08	2E-03	2E-12	2E-03
	Less degraded samples															
	ingestion	7E-03	6E-03	7E-03	6E-03	4E-03	9E-03	6E-02	4E-02	9E-02	1E-04	2E-04	1E-04	3E-02	1E-02	7E-02
HQnc	inhalation	1E-04	1E-04	1E-04	8E-06	1E-05	6E-06	8E-05	5E-05	1E-04	2E-07	1E-07	3E-07	4E-05	9E-06	9E-05
	dermal	8E-03	7E-03	8E-03	2E-01	2E-01	3E-01	1E-02	1E-02	2E-02	2E-07	2E-07	3E-07	2E-02	1E-02	5E-02
	HI_C	1E-04	9E-14	1E-04	3E-06	3E-14	3E-06	2E-05	6E-15	2E-05	4E-07	2E-16	3E-07	8E-02	2E-12	6E-03
children	More degraded samples															
	ingestion	1E-01	4E-02	2E-01	5E-02	3E-02	8E-02	5E-01	3E-01	6E-01	1E-03	9E-04	2E-03	7E-02	5E-02	9E-02
HQnc	inhalation	4E-05	6E-05	3E-04	2E-05	9E-06	3E-05	7E-04	5E-04	9E-04	2E-06	1E-06	2E-06	1E-04	8E-05	1E-04
	dermal	2E-01	8E-02	4E-01	3E+00	2E+00	5E+00	2E-01	1E-01	2E-01	3E-06	3E-06	5E-06	2E-01	1E-01	2E-01
	HI_C	4E-03	9E-14	3E-03	4E-05	2E-12	3E-05	2E-04	4E-15	1E-04	4E-05	4E-15	2E-05	1E-02	8E-12	9E-02
	Less degraded samples															
	ingestion	3E-02	2E-02	3E-02	2E-02	2E-02	4E-02	2E-01	2E-01	4E-01	6E-04	4E-04	8E-04	1E-01	5E-02	3E-02
HQnc	inhalation	2E-04	1E-04	1E-04	8E-06	6E-06	1E-05	4E-04	2E-04	6E-04	8E-07	1E-06	8E-07	2E-04	4E-05	4E-04
	dermal	5E-02	4E-02	6E-02	1E+00	1E+00	2E+00	9E-02	6E-02	1E-01	2E-06	1E-06	2E-06	3E-01	9E-02	7E-01
	HI_C	4E-04	6E-13	3E-04	2E-05	9E-15	3E-06	1E-04	3E-14	6E-05	1E-06	1E-15	9E-07	4E-02	7E-12	3E-02

HIc : Hazard index for Carcinogenic risk **HQnc: Hazard Quotient for non-Carcinogenic risk**

Table 4 POTENTIAL HUMAN HEALTH RISK ASSESSMENT INDEX OF HEAVY METALS OF ACTIVE LANDFILL PRECURSORS (Non-carcinogen and carcinogenic)

Age group	Element	Cu			Cd			Pb			Ag			Zn		
Adult	More degraded samples															
		mean	mini	max	mean	mini	max	mean	mini	max	mean	mini	max	mean	mini	max
	ingestion	4E-03	2E-03	7E-03	5E-03	3E-03	8E-03	4E-02	4E-02	5E-02	2E-04	1E-04	2E-04	6E-03	4E-02	5E-02
HQnc	inhalation	7E-05	4E-05	1E-04	7E-06	1E-05	6E-06	5E-05	5E-05	6E-05	2E-07	2E-07	2E-07	8E-06	1E-05	7E-06
	dermal	5E-03	3E-03	8E-03	2E-01	1E-01	3E-01	1E-02	9E-03	1E-02	3E-07	2E-07	3E-07	8E-06	1E-05	7E-06
	HI_C	1E-04	2E-14	8E-05	3E-06	8E-15	2E-06	1E-05	3E-15	1E-05	3E-07	4E-16	3E-07	6E-04	7E-13	5E-04
	less degraded samples															
	ingestion	3E-03	2E-03	4E-03	3E-03	1E-03	5E-03	3E-02	2E-02	4E-02	2E-04	1E-04	4E-04	4E-03	2E-02	4E-02
HQnc	inhalation	5E-05	3E-05	7E-05	4E-06	2E-06	6E-06	4E-05	2E-05	6E-05	3E-07	1E-07	5E-07	6E-06	3E-06	1E-05
	dermal	3E-03	2E-03	5E-03	1E-01	5E-02	2E-01	6E-03	4E-03	1E-02	4E-07	2E-07	7E-07	6E-06	1E-05	4E-06
	HI_C	6E-05	1E-14	5E-05	2E-06	1E-14	1E-06	5E-06	3E-15	1E-05	2E-07	2E-16	7E-07	9E-04	3E-13	7E-04
Children	More degraded samples															
	ingestion	2E-02	7E-04	7E-04	2E-02	1E-02	3E-02	2E-01	2E-01	2E-01	6E-04	5E-04	7E-04	2E-02	1E-03	4E-02
HQnc	inhalation	2E-05	1E-05	4E-05	3E-05	2E-05	5E-05	3E-04	2E-04	3E-04	1E-06	8E-07	1E-06	4E-05	2E-05	6E-05
	dermal	3E-02	2E-02	5E-02	1E+00	7E-01	2E+00	6E-02	6E-02	7E-02	2E-06	2E-06	2E-06	6E-02	3E-02	9E-02
	HI_C	5E-05	6E-15	2E-04	1E-05	6E-15	7E-06	5E-05	1E-14	6E-06	1E-06	2E-15	2E-06	4E-03	3E-12	5E-03

Less degraded samples

	ingestion	1E-02	7E-03	2E-02	1E-02	1E-02	6E-03	1E-01	2E-01	8E-02	8E-04	4E-04	2E-03	2E-02	8E-03	3E-02
HQnc	inhalation	2E-05	1E-05	2E-05	2E-05	9E-06	3E-05	2E-04	1E-04	3E-04	1E-06	8E-07	2E-06	3E-05	1E-05	5E-05
	dermal	2E-02	1E-02	3E-02	8E-01	3E-01	1E+00	4E-02	3E-02	7E-02	2E-06	1E-06	5E-06	4E-02	2E-02	7E-02
	HI_C	3E-04	3E-13	2E-04	8E-06	8E-15	7E-06	5E-05	6E-15	2E-05	3E-06	4E-15	1E-06	2E-03	5E-12	1E-03

HIc : Hazard index for Carcinogenic risk **HQnc: Hazard Quotient for non-Carcinogenic risk**

Figure

Figure 1

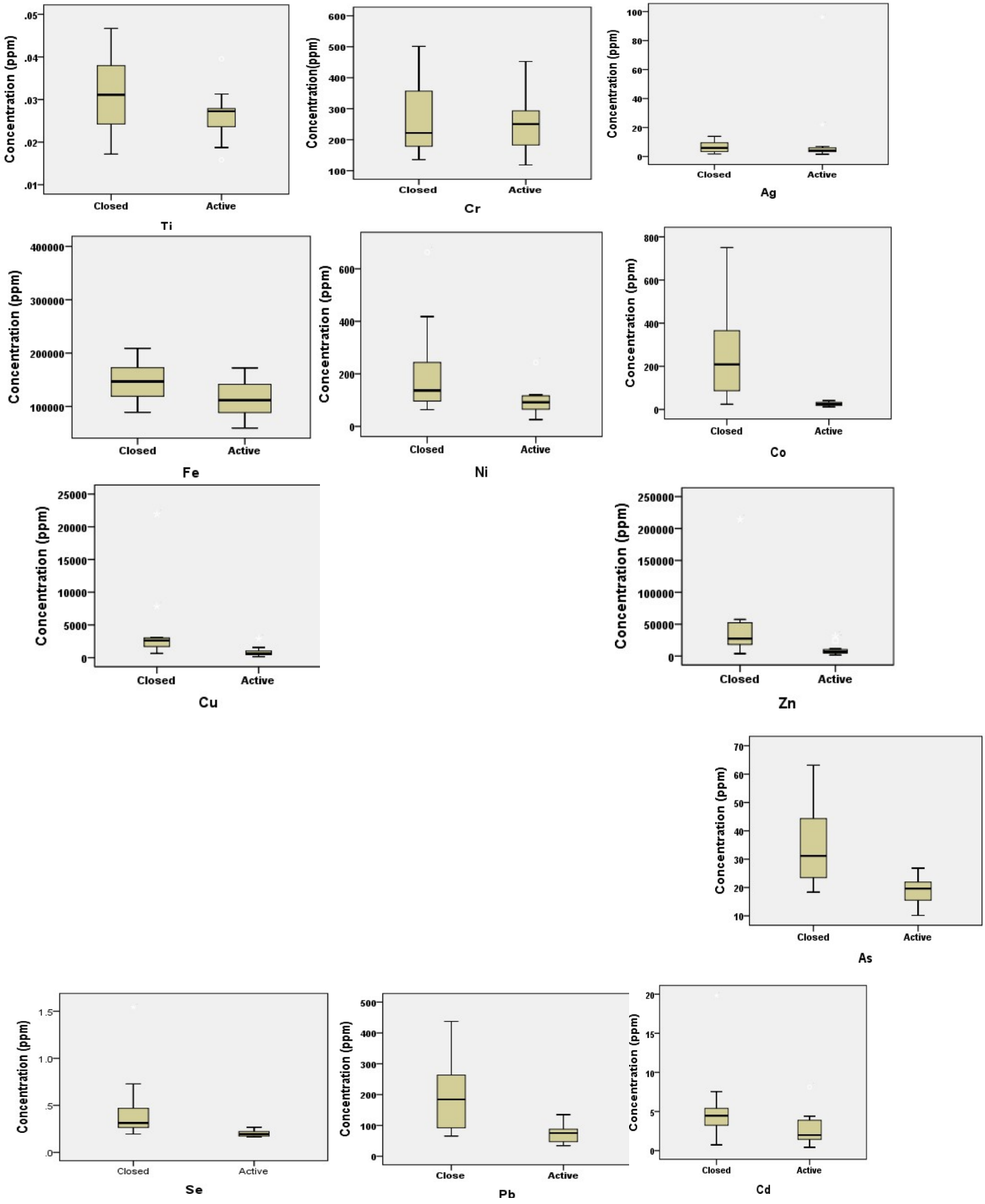
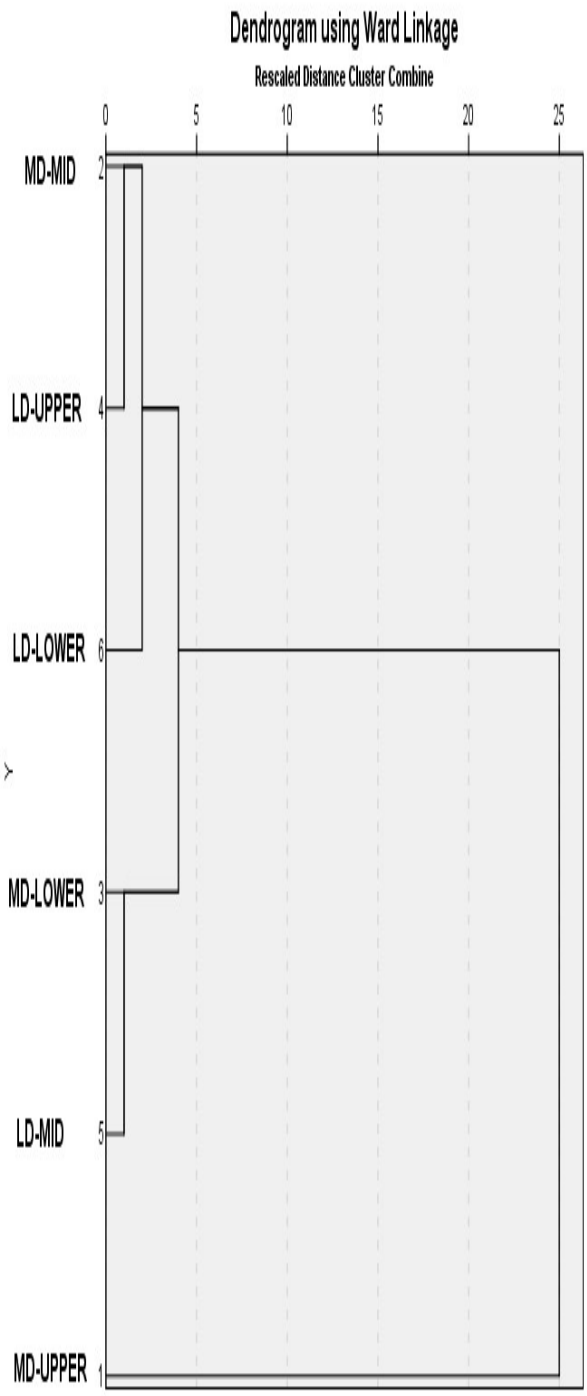
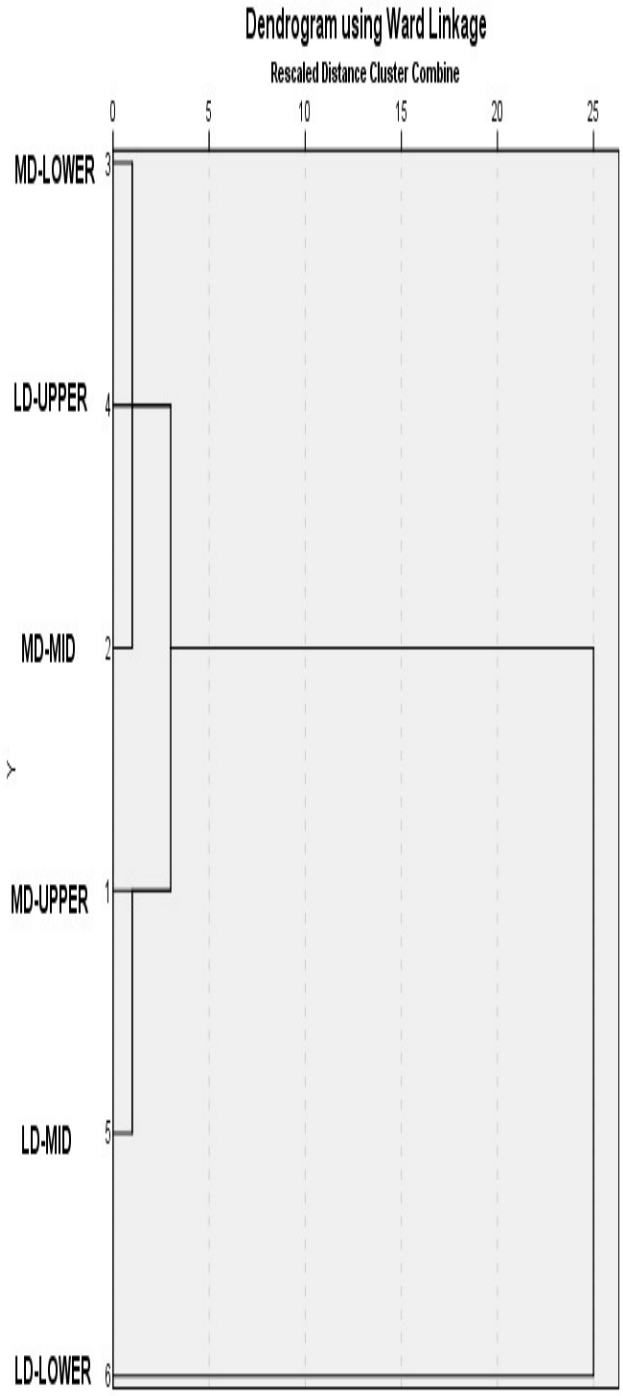

Concentration (ppm)
Closed
Active
Ti
Concentration(ppm)
Cr
Ag
Fe
Ni
Co
Cu
Zn
As
Se
Pb
Close
Cd

Figure 2





Closed Landfill

Active landfill

Figure 3

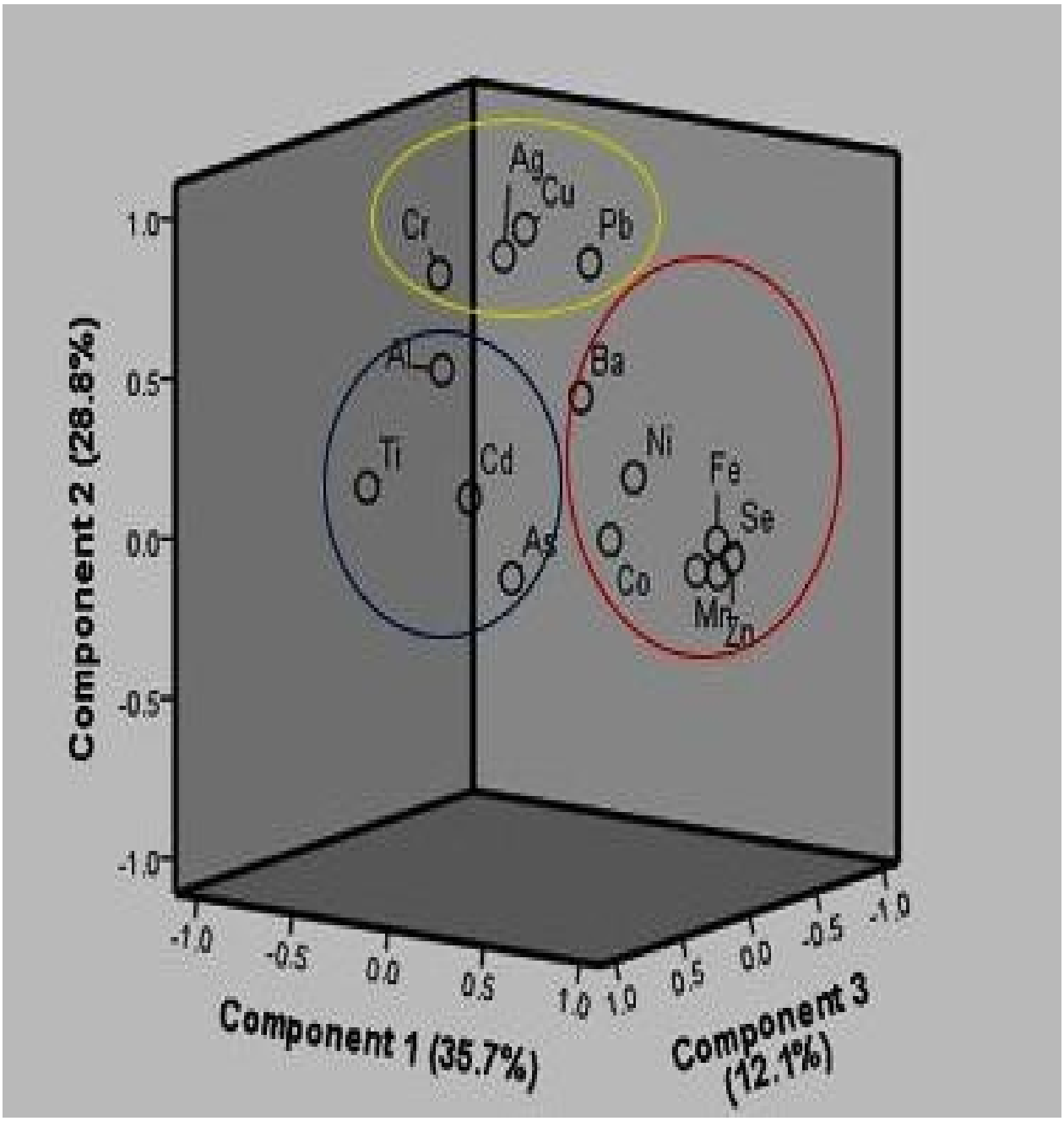

Component 2 (28.8%)
1.0
0.5
0.0
-0.5
-1.0
Ag
Cu
Cr
Pb
Al
Ba
Ti
Cd
Ni
Fe
Se
As
Co
Mn
Zn
-1.0
-0.5
0.0
0.5
1.0
Component 1 (35.7%)
Component 3 (12.1%)

Figure 4

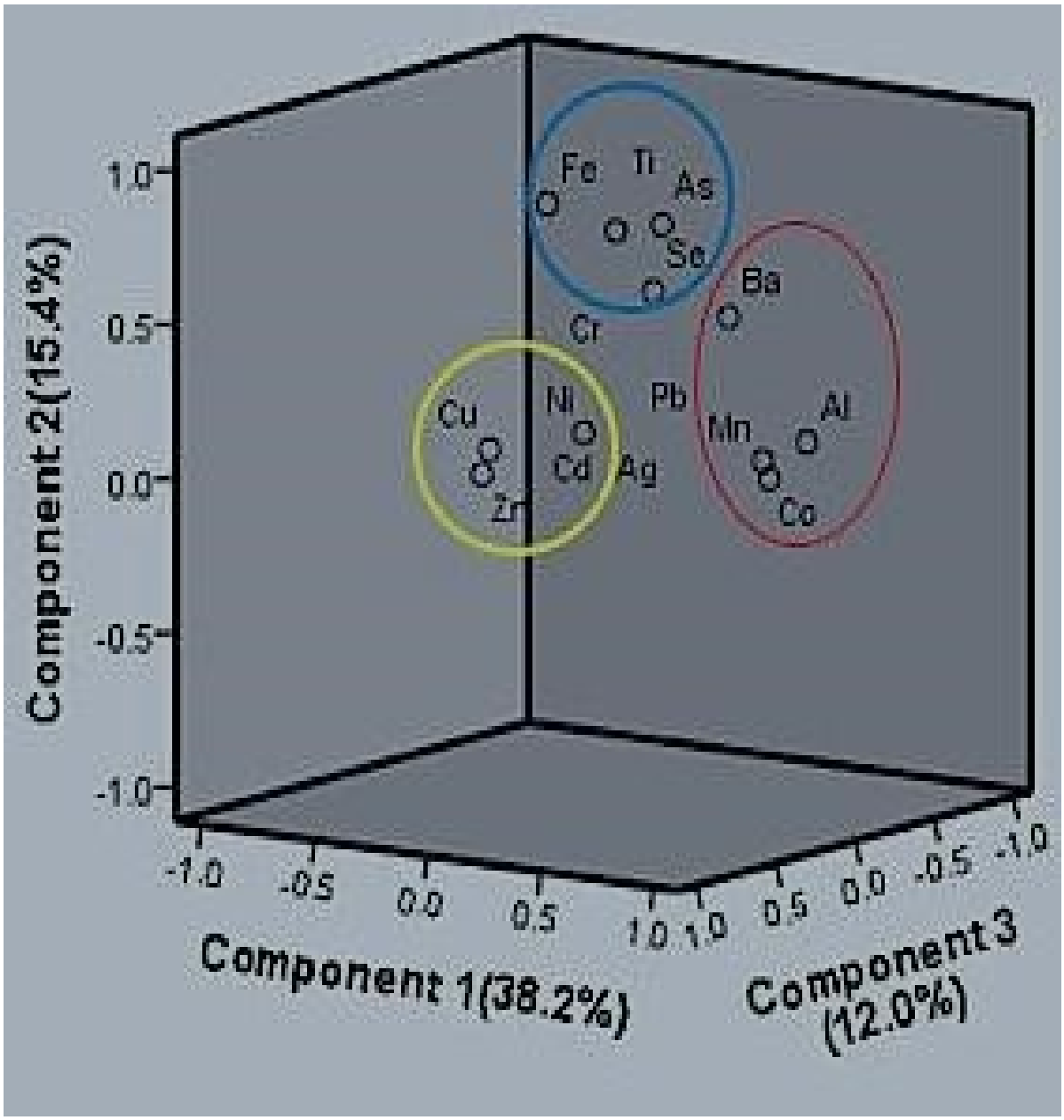

Component 2(15.4%)
1.0
0.5
0.0
-0.5
-1.0
Fe
Ti
As
Se
Ba
Cr
Cu
Ni
Pb
Mn
Al
Cd
Ag
Zn
Co
-1.0
-0.5
0.0
0.5
1.0
Component 1(38.2%)
Component 3
(12.0%)

Figure 5