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Assessment of Trace and Major Elements Contamination in Waste Soils: Leaching Potential from Active and Closed Landfills in Lagos, Nigeria

Akinranti S. Ajibola
Department of Chemical Sciences, Adekunle Ajasin University, Akungba-Akoko, Nigeria

Abstract
Lagos with a population of about 21 million people is the commercial capital of Nigeria. About nine million metric tonnes of wastes were deposited into various landfill sites in Lagos in 2014. Many existing and abandoned landfills pose serious detrimental health impacts to the environment. This study evaluates the trace and major elements (Zn, Pb, Ni, Cd, Cu, Mn, Fe, Ca, K and Na) composition of waste soils from five landfills (three closed and two active landfills) in Lagos and the leaching potential of these elements from waste soils with a view to assessing the potential contamination of the groundwater resources near the landfill sites. The US EPA Toxicity Characteristics Leaching Procedure (TCLP) was employed as the leaching test method. The concentrations of trace and major elements in waste soils were determined. TCLP extracts were also analyzed for the selected trace and major elements. The concentrations of Zn in all the landfill sites (closed and active) exceeded the LASEPA regulatory limit for soil. Worthy of note was the high concentration of Pb in waste soils from Abule-Egba (closed landfill). The concentrations of the analyzed metals in TCLP extracts were found to be lower than the allowable concentrations (especially Cd, Pb and Ni) based on the TCLP standard. With the exception of Zn and Ni in Abule–Egba landfill where a leaching rate of about 24% was observed the leaching rate of trace metals were higher in active landfill sites than the closed landfills. Considering the high leaching rate of Zn and Ni in Abule-Egba landfill and the considerable leaching potential of other trace metals, it is recommended that Abule-Egba landfill, despite its current closure to waste disposal activities, should be more closely monitored in order to prevent possible contamination of groundwater resources around the landfill.
Keywords: Landfill, Solid waste, waste soil, leaching potential, trace and major elements, Lagos

1.Introduction

Attaining environmentally sound management of waste is a problem in many urban and rural areas of most countries of the world. Many areas, particularly in developing countries still have inadequate waste management infrastructure with uncontrolled open dumps and illegal roadside dumps remaining a problem. Such indiscriminate dumping of waste may degrade scenic resources and landscape, pollute soil and water resources, and is a potential health hazard to plants, animals and humans (Baderna et al., 2011). Landfilling is widely applied as a disposal method for municipal solid waste in most developed and developing countries due to such advantages as simple disposal procedure, low cost and landscape restoring effects on pits from mineral workings (Davis and Cornwell, 2008). Non recyclable wastes are often disposed of into landfills. Waste soils generally constitute about 75% by mass of these non-recyclable wastes (USEPA 1997). High levels of heavy metals in these waste soils are extremely toxic to plants (Farrell and Jones 2009).

Globally, many existing landfills pose serious detrimental health impacts to the environment. Degradation of organic wastes dumped on these landfills produces methane which is a green house gas and due to its flammability can be a danger to local inhabitants (Rong et al. 2015, Mou et al. 2014). It has been suggested that abandoned or closed landfill sites could pose as much threats and risks to the neighbouring groundwater and surface water resources as active dumpsites could. In some situations, a closed dumpsite can even pose more threat than an active dumpsite. A study carried out on an abandoned dumpsite supported the assertion that most closed dumpsites in heavily urbanized society may have been contaminated significantly (Adewuyi and Opasina, 2010). A recent study showed that aqua-regia extractable concentrations of heavy metals in soil from a waste landfill were very high though the mobility and risk of leaching was disproportionately low (Gwenzi et al. 2016). Knowledge about the leaching potential of trace and major elements is often required to assess the potential risks of dumpsites to human and environment (Karim et al. 2014). The environmental impacts of toxic substances from the municipal solid waste disposal sites have been increasing at an alarming rate, especially the leaching of heavy metals (Prechthai et al 2008; Iqbal et al. 2015). The environmental problem with heavy metals is that they are unaffected during degradation of organic waste and have toxic effects on living organisms when exceeding a certain concentration. Although some of these elements are necessary for the health of humans in minute amounts (micronutrients like Cu, Mn, Ni, Zn), an excess could be harmful. Some elements are carcinogenic or toxic and may affect the central nervous system (Mn, Pb), the kidneys and liver (Pb, Cd, Cu) or skin, bones and teeth (Ni, Cd, Cu) (Hogan 2010). In most cases, in order to assess the potential health hazard of such elements or the toxicity of the element itself, it is necessary to consider its solubility in a given medium (Fuente-Cuesta et al.

2013).

Leaching tests are often applied in assessing worst case environmental scenario where components of the samples become soluble and mobile. The knowledge of heavy metal contents and leachability from landfill is a pre-requisite for assessing potential hazards to the environment (Karim et al. 2014). The mobility of toxic metals released from wastes is assessed using a variety of risk assessment procedures including the Toxicity Characteristic Leaching Procedure (TCLP), the Synthetic Precipitation Leaching Procedure (SPLP), Multiple Extraction Procedure (MEP), Waste Extraction Test (WET), among others. The TCLP was designed to simulate the leaching of heavy metals and organics from industrial wastes co-disposed in a municipal landfill (Al-Abed et al. 2006).

The current disposal practices of solid wastes in Nigeria are not totally safe for humans and the environment; the waste disposal sites are neither properly designed nor constructed. Assorted solid wastes are indiscriminately disposed at the landfills without segregation. In Nigeria, the major concerns about solid waste management are related to the pollution potential of uncontrolled leachate migration from landfills into local groundwater and the challenge to the attainment of the United Nations' Sustainable Development Goal on clean water and sanitation by 2030. Lagos State is the commercial capital of Nigeria with a population of more than 21 million and an annual growth rate of 3.2 %. Because of the high human population, the state generates a large volume of waste daily which are often disposed of into open dumps or landfills under the management of the Lagos State Waste Management Authority (LAWMA). The landfills receive a wide spectrum of wastes ranging from organic to inorganic and hazardous wastes.

The term 'trace element' is used to designate elements with no known physiological function which, when present in sufficient concentrations, may be toxic to living systems. Other terms used in place of 'trace elements' are 'trace metals' and 'heavy metals'. The use of the term 'heavy metals' is usually, but not always, restricted to those metals with densities greater than 5.0 g cm^{-3}. Trace elements are defined in this study as those elements having less than 0.1 % average abundance in the earth's crust (Bradford et al. 1996). Using this definition the elements Ca, Fe, K, and Na are considered 'major' elements in this study while Zn, Pb, Cd, Cu, Ni and Mn are considered trace elements.

The objectives of this study were to evaluate and compare the trace and major elements composition of waste soils from three closed and two active dumpsites in Lagos, Nigeria and to determine the quality characteristics of simulated leachates prepared from waste soils obtained from the active and closed landfill sites in order to assess the leaching potential of trace and major elements pollutants into the surrounding groundwater and surface water resources of the dumpsites. In this regard, waste soil samples were collected from five dumpsites, sorted, characterized and analyzed for some trace and major elements. The leaching potential of pollutants (trace and major elements) from the waste soils was also evaluated by using the Toxicity Characteristics Leaching Procedure (TCLP) test method.

2. Materials and Methods

2.1 Sites Description:

Olusoshun, Abule-Egba and Soluos landfills are located in Lagos, South-Western Nigeria (Fig. 1). Lagos is located on longitude 3º 24' E and latitude 6º 27' N. Five landfill sites (three closed and two active sites) under the management of the Lagos Waste Management Authority (LAWMA) were considered for this study: Olusoshun active site (OL), Olusoshun closed site (OLC), Soluos 2 closed site (S2,) Soluos 3 active site (S3) and Abule-Egba closed site (AE). Olushosun landfill is located at Ojota, Lagos State, Nigeria. It began operation in 1992. The size of the landfill is about 42.7 hectares. Therefore, the lifespan of the landfill is 24 years. Waste deposited on the landfill is predominantly solid wastes from surrounding industrial factories, gasoline station, automobile repair workshop and waste transported to the landfill. The landfill shares same boundary with Oregun, Ketu and Ojota communities. It receives approximately 40% of the total wastes deposits in Lagos. Solous landfill is ranked the second largest after Olusoshun dumpsite. Solous landfill is sub-divided into two sections namely Solous 2 (closed site) and Solous 3 (an active site). Solous 2 covered about 7.8 hectares with average lifespan of 5 years and Solous 3 covered about 5 hectares of land with an average life span of 5 years and receives an average waste of about 2,250m^2 per day (LAWMA website). Abule-Egba landfill, which began operation in 1992, is located along Oshodi–Sango road in the Alimosho Local Government area, in the North Western part of Lagos State, Nigeria. The land size is about 10.2 hectares with a lifespan of about 24 years from the date of its establishment in 1992. Its geographical location is 6.87ºN, 3.38ºE. It received 250,000 tonnes of waste annually. Abule-Egba landfill has been inactive (closed) since 2008.

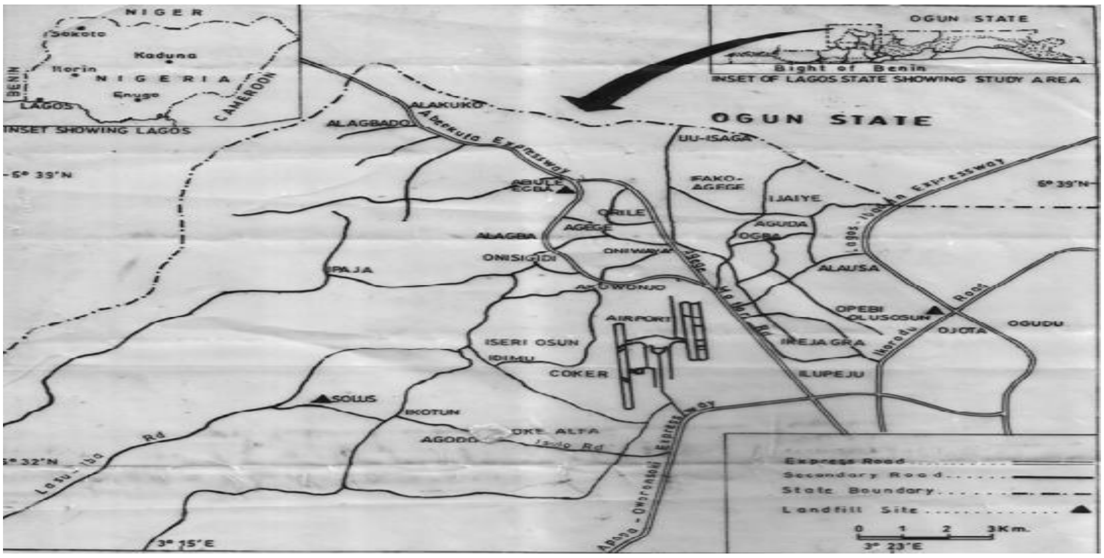

Fig 1: Map extract of Lagos showing the location of the Landfill Sites

2.2 Sampling and Sample Preparation:
Waste soil samples were collected from the dumpsites in July 2015. The soil samples were collected beneath the dumpsites at a depth of between 5 and 15 cm. Waste soil samples of about 6 kg were collected from each landfill sites. The samples were air-dried, sorted and homogenized.

2.3 TCLP Extraction:
The leaching potential of trace and major elements in the waste soils was determined according to the Toxic Characteristic Leaching Procedure (TCLP) test method. Based on this extraction procedure, the extract solution with a pH of 4.93±0.05 was used for this test. The extraction fluid was prepared by the addition of 64.3mL of 1N NaOH solution to 5.7mL acetic acid and then adjusted to 1000mL with distilled water. This test was performed in duplicates. The sample was prepared by mixing of waste soil sample and extraction fluid in the ratio of 1:20 (solid: extraction fluid) in a vessel. The sample was then agitated at 30 rpm for 18 hours. For each sample preparation, a 50g of waste soil sample was mixed with 1000 mL of extraction fluid, and agitated for 18 hours. The extract was filtered, poured into in a 1L plastic bottle and stored at 4ºC until analysis.

2.4 Laboratory Analysis: Total concentrations of trace and major elements in waste soils were determined by digestion using aqua-regia (a mixture of 70% HNO_3 and HCl (1/3 v/v). 0.5 g of waste soil sample was digested by adding 20 mL of aqua-regia, heated to about temperature of 90ºC and then filtered. TCLP extracts (leachates) were digested with concentrated HNO_3. Analysis for trace and major elements in both the waste soils and leachates was carried out using an Atomic Absorption Spectrometer (Thermo Fisher Scientific iCE 3000 Series). The analyses were performed in triplicates.

3. Results and Discussion
3.1 Trace and Major Elements Composition of Waste Soils:
The average concentrations of trace and major elements in the waste soils are shown in Figures 2 and 3. The trace elements determined include Zn, Pb, Cu, Ni, Cd, Mn while the major elements include Ca, Na, Fe and K. The concentration level of each of the trace and major elements in the waste soils from the landfills was compared with the Lagos Environmental Protection Agency (LASEPA) regulatory standards where available.
In general, the closed landfill sites had higher levels of Zinc than the active landfill sites. Abule-Egba landfill contained the highest Zn concentration. High concentrations in waste soils from closed landfill sites may be attributed to high inorganic contents. Similar concentrations of Zn in waste soils from a closed dumpsite were reported by Karim and co-workers (Karim et al 2014). High concentration of Zn in waste soils from a closed irregular landfill has also been reported (Rong et. al 2015). The concentrations of Zn in all the landfill sites (closed and active) exceeded the LASEPA regulatory limit for soil. The presence of Zn at these high levels in waste soils from the landfills calls for concern as high zinc intake beyond permissible limits can produce toxic effects on the immune system. High levels of Zn in soil inhibit many plant metabolic functions resulting in retarded growth. Zn toxicity also causes chlorosis in the younger leaves after a prolonged exposure to high level of Zn (Ebbs and Kochian 1997). The high concentration of Zn in the landfill sites could be as a result of dumping used batteries and fluorescent lamps into the landfills.

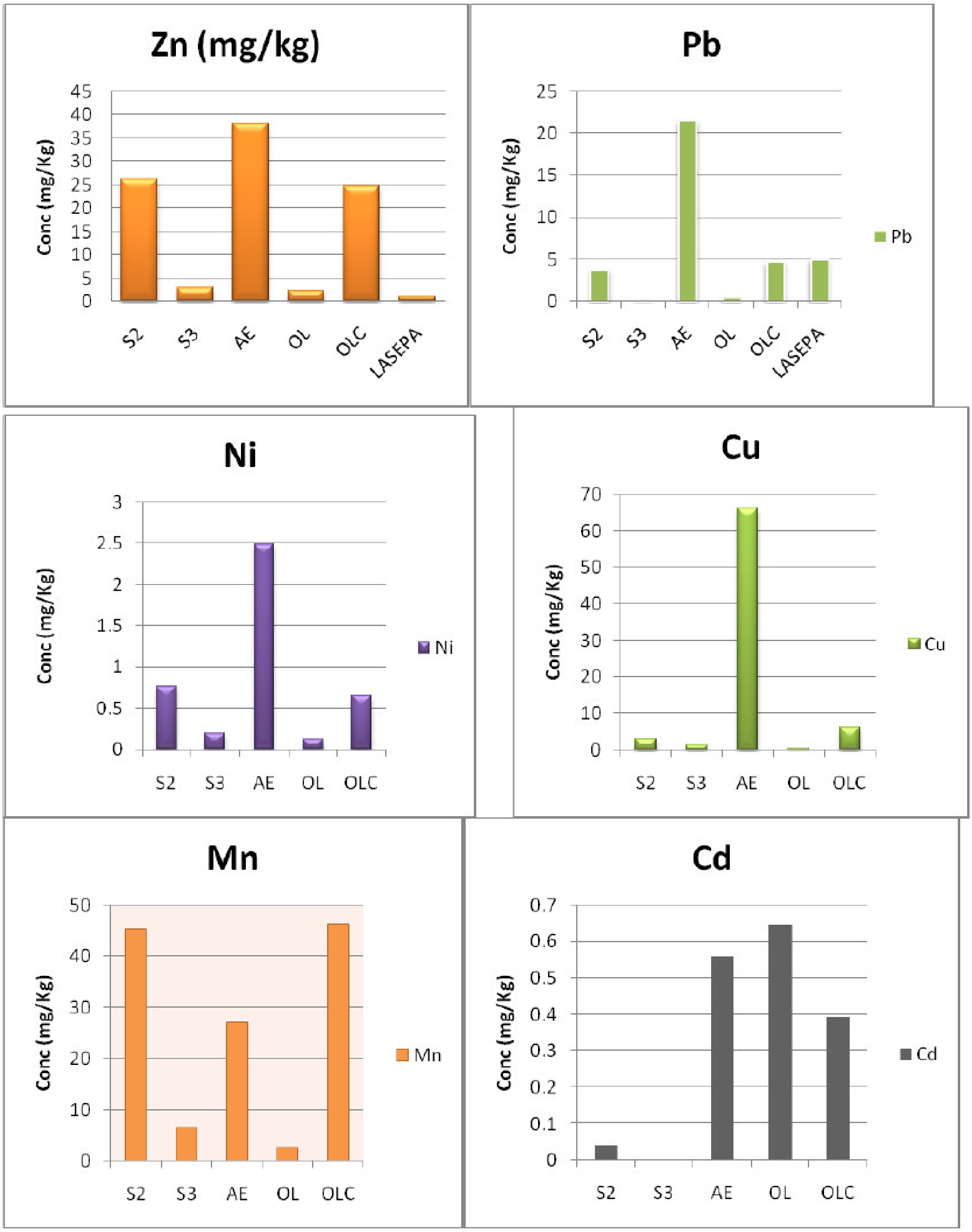

Fig 2: Trace Elements concentrations in waste soils from active and closed landfills

Moreover, the concentrations of Pb in the waste soils from all the landfills were below the LASEPA regulatory limit (5mg/kg) with the exception of Abule-Egba that contained up to 21.5 mg/kg. The closed landfill sites contained higher levels of Pb than the active landfill sites. Similar studies also reported higher levels of Pb in closed dumpsites than closed dumpsites (Karim et al. 2014, Essaku et al. 2008). Worthy of note was the high concentration of Pb in Abule-Egba (closed landfill) which could be attributed to the dumping of Pb-related wastes such as used batteries and photographic processing materials in the landfill (Moturi et al. 2004). The presence of high levels of Pb in the closed dumpsites is a matter of concern as Pb is known to be one of the highly toxic environmental pollutants.

Higher levels of Ni were found in the closed landfill sites than the active landfills. Similar trends were observed for Cu and Mn. Abule-Egba had a considerable higher level of Cu than either of other two closed landfills (S2 and OLC) which indicate that dumping of waste related to cement-like bags was prominent in Abule-Egba landfill before it was closed for disposal of wastes. The concentration level of Cd was significant only in Olusoshun landfills (both active and closed) and Abule-Egba. Cd was not detected in Soluos 3 landfill. The presence of Cd could be attributed to dumping of plastic softeners, stabilizers and pigments (Rotter et al 2004, Rong et al 2015).

Regarding the major elements, the concentrations of Fe in all the waste soils exceeded the LASEPA permissible limit (as shown in Fig. 3) indicating that the waste soils were contaminated with Fe. The high level of Fe in the waste soils is an evidence of dumping of iron and steel scraps wastes in the landfill sites. Though all the landfill sites contained similar levels of Fe, the concentration of Fe were higher in active sites than the closed landfill sites. However, Ca contents in waste soils from all the landfill sites were within the LASEPA regulatory limits as shown in Fig. 3. Waste soils in all the landfill sites contained similar value of Ca contents with the exception of Soluos 2 and Olusoshun active site (OL) where Ca was not detected. Na was detected in Soluos landfill sites (active and closed) and Abule-Egba but was not detected in Olusoshun landfill sites (active and closed). However, K was detected in waste soils from all the landfills with Soluos 3 containing the highest level.

The results of the analysis of waste soils for trace and major elements shows that the closed landfill sites (especially Abule-Egba closed landfill) were contaminated with most metals determined in this study. This further supports the assertion that a closed (abandoned) dumpsite could be more contaminated than an existing dumpsite (Adewuyi and Olapade 2010).

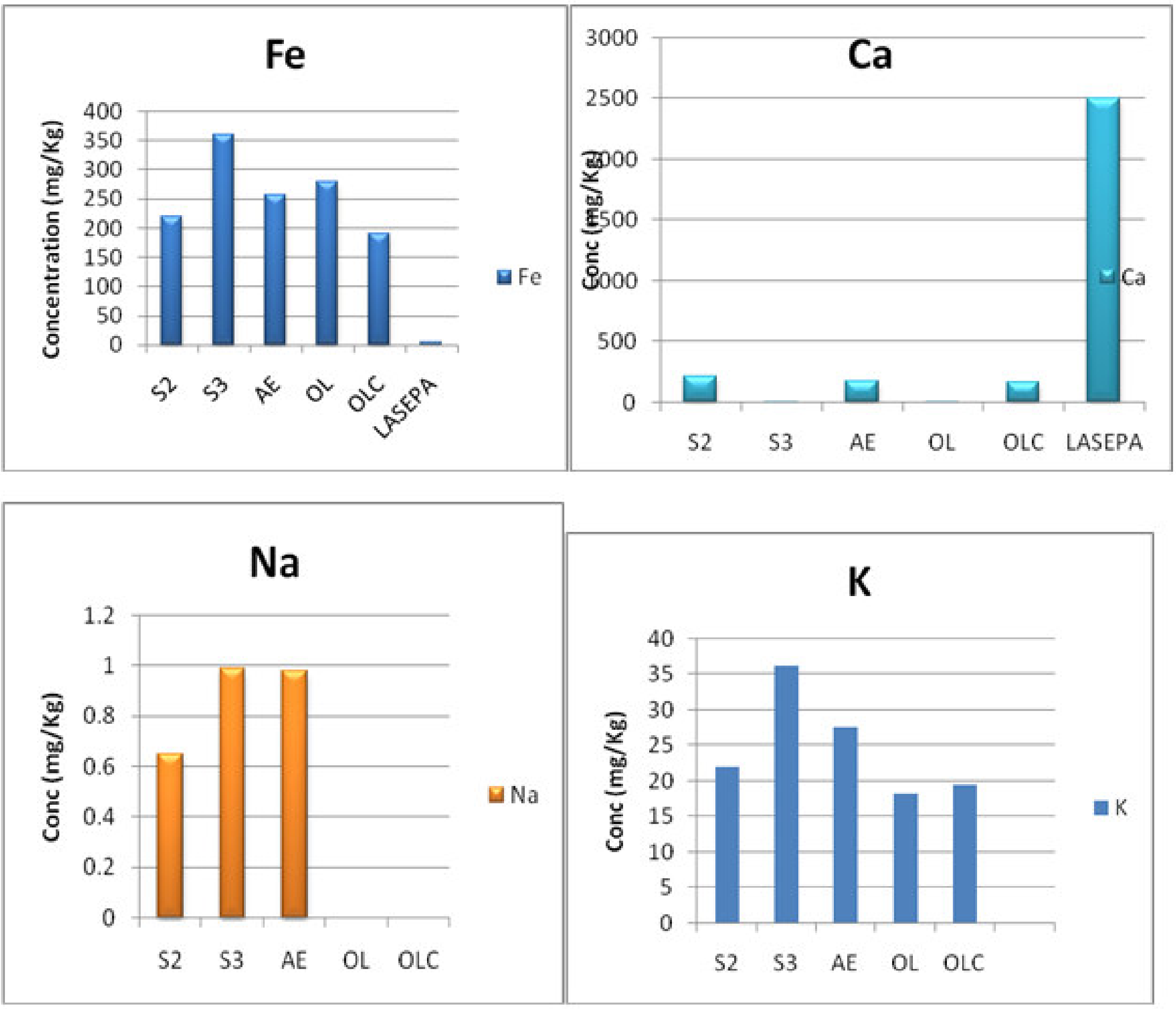

Fig 3: Major elements concentrations in waste soils from active and closed landfills

3.2 Leaching Potentials of Trace and Major Elements in Waste Soils

The quality characteristics of extracted leachates from the active and closed landfills are presented in Table 1. The pH values of the extracted leachates ranged from 4.9 to 6.2 with the closed landfills having the highest pH values. The acidic values were due to the nature of the extractants (acetic acid) employed by the TCLP. The slight difference in the pH values of the closed and active landfills was due to the degree of acid buffer capacity of the waste soils to neutralize the pH of extracted leachates (Prechthai et al. 2008). Acidic environment can enhance the mineralization of waste soils which on the other hand led to high Ca contents in the extracted leachates from all the waste soils. The concentration of Zn in the extracted leachates ranged from 36 to 439 µg/L. Highest concentration of Zn was observed for Abule-Egba dumpsite and this is a reflection of its high concentration found in the corresponding waste soil from the dumpsite (Fig 2). High concentration of Zn in TCLP extracted leachate was also observed by Karim et. al 2014. The concentration of Pb was generally low (1.0-12 µg/L). Maximum concentration of Pb (12 µg/L) in extracted leachates was also found in Abule-Egba dumpsite. However, in comparison with the TCLP limit the concentration was well below the limit of 5000 µg/L for Pb. Consequently, waste soils from all the landfill sites may not be classified as hazardous for Pb based on TCLP limit. Ni concentration in the extracted leachates was noticeably high in the two active landfill sites (Soluos 3 and Olushosun) in comparison with the closed dumpsites. Cd contents in the extracted leachates were high though did not exceed the TCLP limit of 1000 µg/L. The concentration of Mn was highest in Soluos 3 despite its low concentration in the corresponding waste soil in comparison with waste soils from other landfill sites. In general, the concentrations of the analyzed metals were found to be lower than the allowable heavy metal concentrations (especially Cd, Pb and Ni) under the US Toxicity Characteristics Leaching Procedure (TCLP) standard (US EPA 2013). Consequently, the risk of metal contamination and environmental hazard associated with the landfill sites may be considered low with respect to Cd, Pb, and Ni contents. A recent study showed that aqua-regia extractable concentrations of heavy metals in soil from a waste landfill were relatively high but the mobility and risk of leaching was disproportionately low (Gwenzi et al. 2016).

For the major elements (Ca, Na and K) the concentration levels in extracted leachates from all the landfill sites were generally high. Ca contents in the extracted leachates ranged from 8711 to 18910µg/L. These values of Ca contents in the extracted leachates are remarkably high indicating a high leaching potential of Ca from the waste soils from all the landfills. Closed landfills contained slightly higher concentrations than the active landfills. Ca ions influence hardness of water thus, groundwater resources around the landfills could adversely be affected with the high leaching potential of Ca. Therefore, groundwater resources around the landfills should be monitored for the presence of Ca.

Table 1: Quality Characteristics of TCLP extracted Leachates from active and closed landfills in Lagos

Parameter	Soluos 2 (closed)	Soluos 3 (active)	Abule-Egba (closed)	Olusoshun (active)	Olusoshun (closed)	TCLP Regulatory Level
pH	5.4±0.8	5.1±1.4	6.2±1.2	5.2±1.6	5.8±0.6	-
Zinc	36±5	159±10	439±25	135±13	52±3	-
Lead	2.0±0.4	4.0±0.5	12.0±2.5	3±0.4	1.0±0.3	5000
Nickel	ND	848±85	29±4	813±92	ND	2000
Copper	ND	13±2	29±4	97±8	6±1	-
Cadmium	ND	715±78	775±84	801±68	755±72	1000
Manganese	170±12	389±32	47±5	175±14	24±3	-
Calcium	10740±964	11322±983	18910±1047	8711±514	15186±1326	-
Sodium	448±35	549±42	725±74	659±50	597±63	-
Potassium	445±41	762±85	775±82	801±71	755±96	-

Note: All values are in mg/L except pH

The leaching rate of the trace and major elements from the waste soil was calculated using Eqs.1 and 2 according to Prechthai et al. 2008.

$$\text{Metal leached (mg/kg)} = \frac{CL \; X \; L}{Sw} \text{ x } 1000 \qquad (1)$$

$$\text{Metal leaching rate (\%)} = \frac{\text{Metal leached}}{Cs} \text{ x } 100 \qquad (2)$$

where *C*L is the trace metal concentration in extracted leachate (mg/L); C_S is the trace metal concentration in waste soil (mg/kg); *L* is the extracted leachate volume (l); S_W is the quantity of waste soil (g). Where the concentration of trace metal is below the detection limit in TCLP extracted leachate 0.001mg/L was used for the calculation. The results of the leaching rate of heavy metals are presented in Table 2.

The leaching rate of Pb was higher in waste soils from active landfills than in closed landfills (up to 60% in Soluos 3). With the exception of Zn and Ni in Abule–Egba landfill where a leaching rate of about 24% was observed the leaching rate of all the heavy metals were higher in active landfill sites than the closed landfills. This may not be unconnected to the fact that under acidic condition heavy metals are much affected and are readily available with relatively high leaching potential (Prechthai et al. 2008; Essaku et al. 2008). Thus, the relatively more acidic solutions of the extracted leachates from the active landfills enhanced the solubility and leaching rate of the heavy metals. However, it should be noted that apart from the pH of extraction medium there are other factors which can influence the leaching potential of metals. Such factors include the particle size of waste soil which determines the surface area exposed to the leaching solution, metalorganic complex formation, oxidation –reduction conditions and contact time. It is suggested that Abule-Ebga landfill despite its current closure to dumping activities should be more closely monitored in order to prevent the high leaching rate of Zn and Ni and considerable leaching potential of other trace metals, prevent possible contamination of groundwater resources near the landfill. Furthermore, the active landfills (Soluos 3 and OL) should be placed on monitoring program so as to forestall any inherent danger that may be associated with the high leaching potential of heavy metals (especially Pb).

Table 2: Heavy metals leaching rate in TCLP leachates

Heavy Metals	Soluos 2 (closed)	Soluos 3 (active)	Abule-Egba (closed)	Olusoshun (active)	Olusoshun (closed)
Zn	2.7	5.51	23	5.72	0.21
Pb	1.1	62	1.1	11.8	0.42
Ni	2.6	-	24	-	3.04
Mn	1.6	6.05	3.47	6.8	1.1
Cu	0.7	19.2	1.6	3.47	1.03

Note: All values are in weight percentage

Conclusion

In this study, the presence of selected trace and major elements in waste soils from five landfills (two active and three closed landfills) in Lagos, Nigeria and possible source of contamination to the surrounding groundwater reserves have been evaluated. The leaching potential of the trace metals was also assessed. The concentrations of Zn in all the landfill sites (closed and active) exceeded the LASEPA regulatory limit for soil. Worthy of note was the high concentration of Pb in Abule-Egba (closed landfill) which could be attributed to the disposal of Pb-related wastes such as used batteries and photographic processing materials in the landfill. The presence of high levels of Pb in the closed dumpsites is a matter of concern as Pb is known to be one of the highly toxic environmental pollutants. The concentrations of Fe in all the waste soils exceeded the LASEPA permissible limit. The concentrations of the analyzed metals were found to be lower than the allowable heavy metal concentrations (especially Cd, Pb and Ni) under the US Toxicity Characteristics Leaching Procedure (TCLP) standard. Based on the high leaching rate of Zn and Ni in Abule-Egba landfill and considerable leaching potential of trace metals, it is recommended that Abule-Egba landfill, despite its current closure to waste disposal activities, should be more closely monitored in order to prevent possible contamination of groundwater resources near the landfill. Finally, considering the high concentrations of Zn and Fe in waste soils from all the landfill sites which exceeded the LASEPA limits, all the landfill sites should be placed on environmental monitoring program for Zn and Fe contents.

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