

Contamination and source identification of the elemental contents of soil samples from municipal and medical waste Dumpsites in Ile-Ife, Nigeria

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Abstract

Contamination in soil samples collected from municipal and medical waste sites was assessed by employing four indices: contamination factor (C_f), degree of contamination (C_{deg}), pollution load index (PLI), and index of geoaccumulation (I_{geo}). The sources of soil contaminants were identified by using Positive Matrix Factorization (PMF). Iron had the highest average concentrations of 46.47 ± 14.03 and 39.42 ± 2.54 $\mu\text{g/g}$ in the municipal and medical waste dumpsites. C_f values were above 6 for both dumpsites, classifying the dumpsite soil as very high contamination with respect to Cr, Fe, Ni, Cu, Zn, As, Cd, and Pb. The overall C_{deg} and PLI values are 176.9 and 170.4 and > 5 for both dumpsite implying very high degrees of contamination and progressive deterioration, respectively. The average I_{geo} values for Zn, Cd, and Pb of the two dumpsites were >3 , indicating that the soil samples at both study areas were classified as highly to moderately polluted. The three identified sources resolved by PMF and their respective percentage contributions were crustal (32 %), scrap metals wastes (40 %), and electronic wastes (28 %). The studied soils were moderately contaminated and this could increase depending on continual waste disposal. An urgent need for solid waste management is recommended in order to safe guard the ecosystem from detrimental impacts of waste and to reduce the human health risks arising from consumption of plants grown around the dumpsites.

Keywords

municipal, medical, waste, contamination, PIXE, PMF

Introduction

In the pre-industrial era, waste generation and disposal were given little attention as populations were small and industrialization was at a minimum. Today, due to the unprecedented increase in populations and uncontrolled urban growth and development, waste generation and disposal have been considered a major public and environmental health issue globally (Amuda et al.,

2014, Ogundele et al., 2018). In Nigeria, the current solid waste management practices are characterized with uncontrolled disposal in the open spaces and at the peripheries of the urban cities. These make it difficult for state and local environmental protection agencies to maintain effective and efficient municipal solid waste management (Olanrewaju and Ilemobade,

2009). Municipal solid waste (MSW) consists of diverse materials generated from domestic, marketplace, commercial, and institutions uses (education and hospital). For example, educational institutions generate waste such as paper, obsolete electronic items and other consumables during teaching and research activities. Hospitals, as a result of demand for health-care services, generate potentially toxic, infectious, and hazardous wastes that are capable of transmitting communicable diseases such as Hepatitis B and HIV/AIDS (Inyang et al., 2013). Also, medical wastes might result from diagnosis, dispensary, treatment and immunization procedures and in research pertaining to the production and/or testing of biomaterials (Cheremisinoff and Shah, 2009).

Open burning in the dumpsite is the oldest and most commonly adopted way of handling solid wastes. However, waste burning under uncontrolled conditions at elevated temperatures yields emissions and residues that contaminate the air, soil, and water bodies around the dump site (Amuda et al., 2014). Incineration provides the most effective and hygienic approach to waste management (Amadi and Nwankwoala, 2013). Combustion in an incineration facility is capital intensive and it requires skilled personnel and careful management, which is likely the reason for its scarcity in developing countries. Globally, only a few educational institutions and, medical/health centers have incineration facilities. Most dumpsites usually serve as a point source of soil pollution. The soil around the dumpsites might be overburdened with both trace and major element contamination. Soil pollution from improper waste disposal and management has received increasing attention in the last few decades in both developing and developed countries (Aelion et al., 2009; Kanmani and Gandhimathi, 2013; Inyang et al., 2013; Sardar et al., 2013; Ogunbanjo et al., 2016; Ogundele et al., 2020a). The contamination of the agricultural soil by municipal and hospital wastes is of serious concern from human health perspectives. An intensive subsistence agricultural practice occurs in close proximity of the dumpsites and their neighborhoods. The possibility of plant uptake of contaminants in the waste is high by the arable crops and animal grazing. Also consumption of such plants and animals by human could increase potential toxicants in human body. Therefore, the present study was conducted to

assess the contamination status of the soil samples from the municipal and medical waste dumpsites

The characterization of soil samples and identification of probable sources of soil contaminants around dumpsites are integral parts of soil reclamation processes (Tariq et al., 2013). The soil characterization (multi-elemental assay) is achieved by employing nuclear and atomic based-analytical techniques (Odukoya, 2015). The measured elemental concentrations from the employed analytical technique usually serve as input data when conducting hazard identification or source identification. In this study, the choice of Particle-Induced X-ray Excitation (PIXE) for the analysis of soil samples collected from the Obafemi Awolowo University campus and Obafemi Awolowo University Teaching Hospital Complex dumpsites in Ile Ife, Nigeria was based on high precision, multi-elemental capacity, simple sample preparation, low detection limit and short time of analysis. The elemental concentrations in soil samples from the two dumpsites were used as input data required for the contamination assessments and source identification studies.

Materials and methods

Sampling and preparation of samples

The soil samples were collected at Obafemi Awolowo University (OAU) campus and Obafemi Awolowo University Teaching Hospitals Complex (OAUTHC) dumpsites, Ile Ife, Nigeria. OAU has municipal waste, while OAUTHC has medical waste. The geology of the study areas forms a complex pattern of the coarse and fine grain gneiss. The soil is mainly derived from materials of the old basement complex and characterized with well drained, medium to coarse textured grain and overlying with weathered rock materials derived from coarse-grained granitic rocks metamorphose sedimentary rock and gneisses (Olorunfemi and Okhue, 1992; Adetunji et al., 2018). The maps of the study locations showing the evenly distribution of the sampling points and the cross sectional views of the two dumpsites are presented in Figure 1 and 2. Each dumpsite covers about six hectares of bare expanse of land with flat terrain. At each site, soil samples from the dumpsites were conducted in an evenly distributed pattern at 0–10 cm using depth calibrated dutch hand soil auger. The samples were collected from the

decomposed portion of the wastes which had been accumulated over a long period of time. The Dutch hand soil auger was cleansed with distilled water and acetone before and after each sampling to avoid cross contamination. For each sample, four subsamples were taken within 1 m² area, then mixed thoroughly to achieve homogeneity before a representative sample was obtained. Forty samples were collected from each site, duly coded, and transported to the laboratory for further preparation and analysis. Before elemental analysis, the soil samples were air-dried at ambient temperature.

Small pieces of plastics, nylon, other small metallic items, coarse debris, pebbles and stones were removed from the samples before sieving. The dried samples were passed through a 2-mm sieve mesh to remove coarse material and debris and homogenized by grinding using an agate mortar and pestle. From each sample, 13 mm diameter and 1 mm thick triplicate pellets were made using a CARVER model manual pelletizing hydraulic machine by applying 12 tons pressure. All these were carried out in the clean environment in the laboratory to avoid contamination of samples.

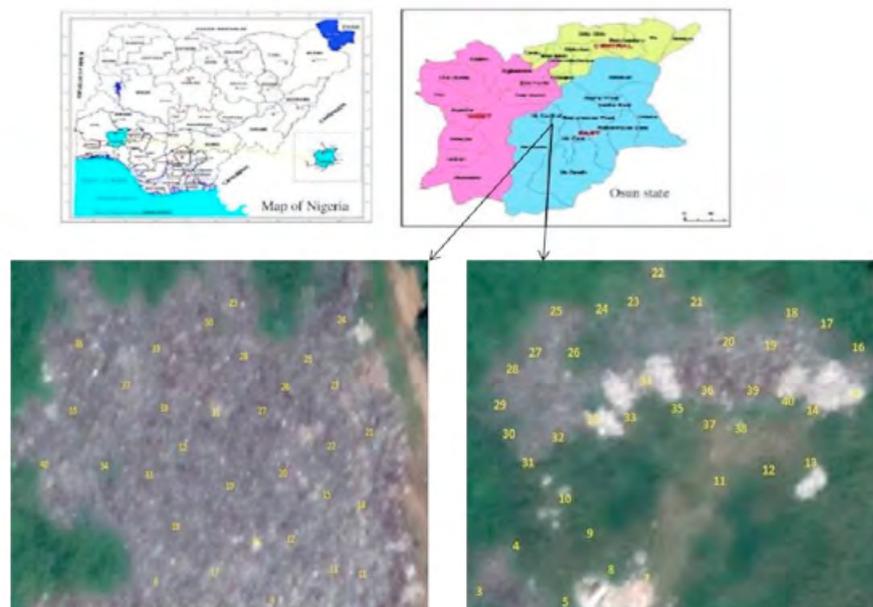


Figure 1. The Map of Nigeria, Osun state (upper panels) and the sampling distributions (lower panels).



Figure 2. Sections of OAU (a) and OAUTHC (b) dumpsites showing waste burning.

Elemental analysis

An Ion Beam Analysis (IBA) facility with a radio frequency (RF) charge exchange ion source provided 2.5 MeV (NEC 5SDH) proton beam for the PIXE analysis. The pelletized samples were bombarded with proton beam from the PIXE channel of the IBA facility. The measurements were performed with a beam spot of 4 mm in diameter and a low beam current of 3-6 nA. A Canberra Si(Li) detector model (ESLX 30-150), beryllium thickness of 25 μm , with energy resolution of 150 eV full width at half maximum (FWHM) at 5.9 keV with the associated pulse processing electronics, and a Canberra Genie 2000 (3.1) multichannel analysis card interfaced to a PC were used for the x-rays data acquisition for a live time of 1000 s. With respect to the beam direction, the sample was placed perpendicular to the beam while the Si(Li) detector was positioned at 45° . The computer code GUPIXWIN 2.2.0 was used for the analysis of the PIXE spectral data. The detail analytical set up and calibration of PIXE facility had been reported elsewhere (Inyang et al., 2012; Olise et al., 2010a). The elemental content of International Atomic Energy Agency (IAEA) Soil 7 Standard Reference Material (SRM) was also determined under the same experimental conditions of PIXE analysis as analytical quality control samples to validate the procedures. The detection limit (DL) of PIXE technique was obtained as $DL = (3.3 * SD)/S$, where SD and S are the standard deviation of the measured blank samples and the slope of the analytical calibration curve for each element (Shrivastava and Gupta, 2011). More details about the analytical set up and procedures had been reported (Ezeh et al., 2018). The PIXE analysis was performed at the Center for Energy Research and Development (CERD), Obafemi Awolowo University, Ile-Ife, Nigeria.

Soil contamination assessment

To assess the contamination status of the soil samples, the contamination factor (C_f), degree of contamination (C_{deg}), pollution load index (PLI) and index of geoaccumulation (I_{geo}) were employed. The determination of each indices are briefly discussed and the categories of values with their associated interpretations for each indices are presented in Table 1. The contamination factor (C_f) for each measured elements in the soil samples was calculated

using the equation below:

$$C_f = \frac{C_s}{C_b} \quad [1]$$

where C_s is the measured elemental concentrations in the soil samples and C_b is the background values of the elements in the parent material (Wedepohl, 1995). The Degree of Contamination (C_{deg}) shows the extent to which the soil of the study areas had been polluted (Ogundele et al., 2020a). It is determined as: The Degree of Contamination (C_{deg}) shows the extent to which the soil of the study areas had been polluted. It is determined as the sum of the individual C_f of all the measured elements:

$$C_{deg} = \sum_{i=1}^n \left(\frac{C_s}{C_b} \right) \quad [2]$$

The Pollution Load Index (PLI) is an empirical index which reveals the extent of cumulative soil pollution load of the measured elements (Gope et al., 2017). PLI is calculated as the n^{th} root of the product of the C_f of the respective elements found in the soil samples. The PLI for the entire site is obtained using equation [3] (Mohiuddin et al., 2010; Ogundele et al., 2017):

$$PLI = \sqrt[n]{C_{f1} + C_{f2} + C_{f3} + C_{f4} + \dots + C_{fn}} \quad [3]$$

where n is the number of the elements under consideration and $C_{f1..n}$ are the contamination factor calculated previously.

Index of Geoaccumulation (I_{geo}) was proposed by Muller (1979) and it had widely employed for determining the soil contamination status. It is computed following the logarithmic function of ratio of measured concentration of the element in the soil (C_n) and geochemical background concentrations (C_b):

$$I_{geo} = \log_2 \left[\frac{C_n}{1.5C_b} \right] \quad [4]$$

In the present study, Wedepohl (1995) continental crustal average data was chosen as the background concentrations for calculating I_{geo} values. The constant 1.5 is introduced to minimize the effect of possible fluctuation in the background values that may be attributed to lithogenic variations in the soil.

Table 1. Pollution indices, categories and interpretation.

Index	Values	Interpretation	References
CF	CF < 1	low contamination	Tomlinson et al. (1980); Seshan et al. (2010)
	1 ≤ CF ≤ 3	moderate contamination	
	3 ≤ CF ≤ 6	very high contamination	
C _{deg}	< 8	low contamination	Tomlinson et al. (1980), Ogundele et al., (2017)
	8-16	moderate contamination	
	16-32	considerable contamination	
	> 32	very high degree of contamination	
PLI	PLI < 1	unpolluted condition	Mohiuddin et al. (2010); Ogundele et al. (2017)
	PLI = 1	baseline level	
I _{geo}	I _{geo} ≤ 0	Unpolluted	Nowrouzi and Pourkhabbaz (2014); Gupta et al., 2014 Ogundele et al. (2019)
	0 < I _{geo} ≤ 1	unpolluted to moderately polluted	
	1 < I _{geo} ≤ 2	moderately polluted	
	2 < I _{geo} ≤ 3	moderately to high polluted	
	3 < I _{geo} ≤ 4	highly polluted	
	4 < I _{geo} ≤ 5	highly to extremely polluted	
	I _{geo} > 5	extremely polluted	

Positive Matrix Factorization

To identify and apportion sources of elemental contents of the soil samples from OAU and OAUTHC dumpsites were merged to provide a sufficiently large data size which is required for PMF model run. The combination of the data was deemed reasonable since waste compositions of the two dumpsites reflect similar characteristics. Briefly, Positive Matrix Factorization (PMF) decomposes the elemental data into factor profiles (mixture of tracers in each profile) and factor contributions (percentage of each tracer in each factor). A conventional mathematical equation for PMF model can be written as:

$$X = GF + E \tag{5}$$

where X is the $n \times m$ matrix of elemental concentrations, G is the $n \times p$ matrix of source contributions, F is the $p \times m$ matrix of source profiles, and E is the matrix of residuals not fitted by the model and is defined as:

$$e_{ij} = x_{ij} - \sum_{k=1}^p f_{ik} g_{kj} \tag{6}$$

where $i = 1; 2, \dots; n$ samples; $j = 1; 2, \dots; m$ elements; $k = 1; \dots; p$ sources.

The PMF minimizes the objective function, Q , based on the uncertainties inherent in the each measured data as:

$$Q = \sum_{i=1}^m \sum_{j=1}^n \left[\frac{e_{ij}}{u_{ij}} \right]^2 \tag{7}$$

The uncertainty matrix, u_{ij} , was estimated using equation-based approach as proposed in the PMF user's guide (Norris et al., 2014):

$$u_{ij} = \sqrt{(e_f * C)^2 + (DL)^2} \tag{8}$$

where e_f , C and DL are error fraction, measured concentration and detection limit of the measured elements, respectively. The error fractions were defined as 5% of the measured concentration value (Norris et al., 2014; Ogundele et al., 2020). The elemental concentrations and the associated uncertainties data served as input for the PMF model run. The PMF software (Version 5.0) was downloaded freely from the United States Environmental Protection Agency (USEPA) website (www.epa.gov). Detailed methodology for source apportionment can be found elsewhere (Norris et al. 2014; Ogundele et al. 2016; Hopke, 2016). The

sources were identified by examining the mixture of specific tracers in each factor profile and the confirmed based on their percentage contributions.

Results and discussion

PIXE analysis results for IAEA Soil-7 SRM ($\mu\text{g/g}$)

Table 2 shows the results of the certified and measured values of the elemental concentrations of IAEA SOIL-7 SRM for PIXE analysis. The measured values sufficiently compared well with the certified values for the respective elements (K, Ca, Ti, Cr, Fe, Cu, Mn, Zn and Zr). A number of elements were about 2-5 % underestimated including Ca, K, Ti, Mn, Zn and Zr. The possible sources of underestimation might be caused by the counting statistics and self-absorption of x-rays problems (Olise et al., 2010b). The measured values were in good agreement with the certified values, providing strong support for the validity of the PIXE analytical procedure used in this study.

Table 2. Elemental concentrations of IAEA Soil-7 standard reference material ($\mu\text{g/g}$).

Elements	Certified Value	Measured Value
K	12100±350	12120±268
Ca	163000±260	163100±204
Ti	3000±64	3004±40
Cr	2±0.1	1.6±0.6
Fe	25700±110	25700±140
Cu	11±0.2	10±0.2
Mn	631±15	612±17
Zn	104±5	103±6
Zr	185±14	184 ±15

Elemental concentration results of the soil samples from the OAU and OAUTHC dumpsites

The average elemental concentrations (\pm standard deviation) quantified by PIXE spectroscopy are presented in Table 3. The standard deviation (\pm SD) described the variation of the concentration around the average values for each measured element. The composition of the studied soil reflects several elements including Ca, K, Ti, Fe, Cd, Pb, Cr and Ni that are routinely measured in environmental sample matrices due to their significant tracers in source identification and apportionment studies (Begum et al., 2010). The PIXE elemental concentrations result across the entire energy spectral has offered this opportunity. The average

concentrations of Fe were 46.47 ± 14.03 and $39.42 \pm 2.54 \mu\text{g/g}$ for municipal and medical waste dumpsite, respectively. The high concentrations of Fe might be linked to its natural abundance and dumping of ferrous metal wastes at varying degree of degradation in the dumpsites. Potassium had the concentrations of 20.08 ± 6.24 and $16.55 \pm 2.4 \mu\text{g/g}$ in OAU and OAUTHC dumpsites, respectively. Apart from contribution from lithogenic sources, waste burning activities could increase the concentration of K in the soil of the dumpsite. The average concentrations of Pb (8.23 ± 1.40 and $9.82 \pm 2.32 \mu\text{g/g}$) were less than $19.0 \mu\text{g/g}$ in an uncontaminated soils (Moshen and Alireza, 2014) while the average concentrations of Cd (4.92 ± 1.24 and 6.54 ± 4.49) were greater than $0.35 \mu\text{g/g}$ for an uncontaminated soil. The elemental concentration in the soil samples could also be influenced by the geology of the parent materials. The varying elemental contents imply anthropogenic influence on the soil. Generally, the concentrations of anthropogenic elements were found to be higher than crustal elements.

Table 3. Average (\pm SD) elemental concentration results ($\mu\text{g/g}$).

Elements	OAU (n=40)	OAUTHC (n=40)
K	20.08 ± 6.24	16.55 ± 2.4
Ca	7.50 ± 2.23	5.34 ± 1.63
Sc	4.23 ± 2.64	5.83 ± 6.86
Ti	4.09 ± 1.87	4.75 ± 1.10
Cr	5.10 ± 1.26	8.80 ± 5.56
Mn	12.20 ± 2.54	17.75 ± 2.11
Fe	46.47 ± 14.03	39.42 ± 2.54
Ni	8.88 ± 3.50	9.62 ± 4.93
Cu	9.86 ± 1.78	12.85 ± 3.95
Zn	16.49 ± 2.57	19.06 ± 2.63
As	4.35 ± 1.77	3.01 ± 1.41
Cd	4.92 ± 1.24	6.54 ± 4.49
Pb	8.23 ± 1.40	9.82 ± 2.32

Contamination assessment results

Figure 3 shows the average values of the contamination factors of the measured elements in the studied soil of the two dumpsites. The average C_f values for the measured elements in the soil samples are $1 < C_f < 3$ for K, Ca, Sc, Al, Ti and Si for both sites, signifying moderate contamination. Elements like Cr, Mn, Fe, Ni, Cu, Zn, As, Cd and Pb had the average C_f values far greater than the upper limit value of 6 for the contamination factor classification.

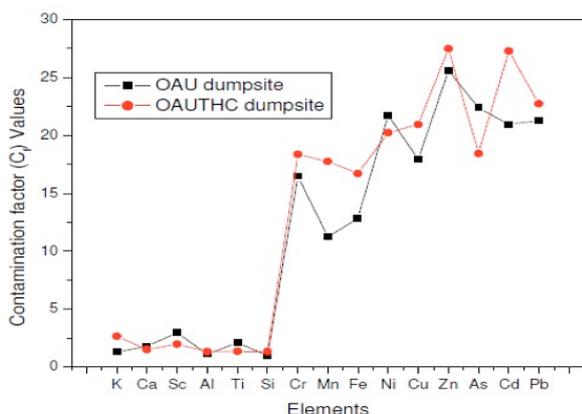


Figure 3. Contamination factors of some measured trace elements for both OAU (municipal dumpsite) and OAUTHC (medical waste dumpsite).

The sequences of Cf values were Zn > As > Ni > Pb > Cd > Cu > Cr > Fe > Mn and Zn > Cd > Pb > Cu > Ni > As > Cr > Mn > Fe for OAU and OAUTHC dumpsites, respectively.

The overall degree of contamination (Cdeg) computed for the OAU and OAUTHC dumpsites were 176.9 and 170.4, respectively. These values were 5.5 and 5.3 times the upper class limit for the Cdeg categories. Based on the overall Cdeg, the two dumpsites were classified as having very high degrees of contamination. The estimated PLI values of the measured elements for OAU and OAUTHC dumpsites are 5.9 and 5.8. These values are much larger than 1, the upper limit of the PLI classification, showing a progressive deterioration of the soil of the dumpsites.

The calculated average Igeo values of the measured elements in the dumpsites soil samples are presented in Figure 4.

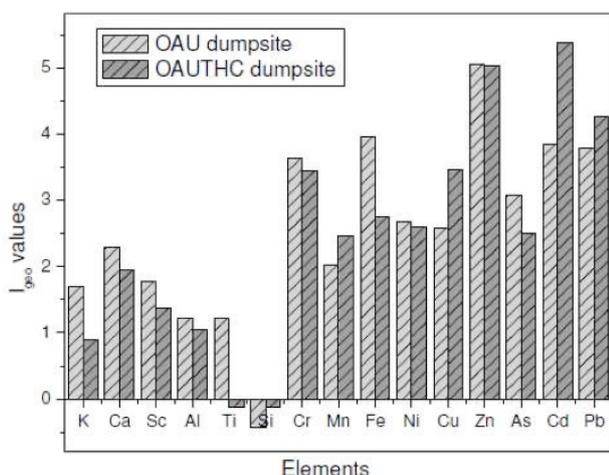


Figure 4. Average values of Index of Geoaccumulation (Igeo) of measured concentration of the element in the soil.

For OAU dumpsite, the average I_{geo} values for the crustal elements (K, Ca, Sc, Al and Ti) were less than 2 (<2), which ranked the dumpsite soil samples as having moderate contamination. This moderate contamination could be as a result of construction/demolition wastes as well weathering of the parent material of the soil. The OAU dumpsite is located in low terrain and accumulation of crustal materials might be high during rainy season. Also, most anthropogenically enriched elements had the average I_{geo} greater than 2 (>2) with order of the average values as follows: Zn < Cd < Fe < Pb < Cr < As < Cu < Mn. The average I_{geo} values for Zn was 5.1, indicating that the soil sample of the study area are classified as extremely polluted with respect to Zn. Also, Fe, As, Cd, Pb and Cr had $3 < I_{geo} < 4$ as the average I_{geo} , which was ranked as heavily to extremely contaminated. The average I_{geo} values of the studied soil of the OAUTHC dumpsite followed similar trends with little variation in both crustal and anthropogenically enhanced elements. The level of contamination by the anthropogenic elements most especially Zn and Cd might be attributed increase accumulated restrained disposal of old galvanized metal sheet and Cd rich waste material in the dumpsite. The importance of cadmium contamination in soils has been emphasized most recently due to its toxicity effects when it enters the human body via food chain (Odukoya et al., 2015).

PMF results

Figure 5 shows the factor profiles resolved by PMF for the combined PIXE elemental concentrations data of the dumpsites. The blue bars indicate the concentrations of the elements in the source profiles while the red square box in each bar denote the percentage of explained variation for that element for that factor.

The left Y-axis is a logarithmic scale correspond mass bar while the right Y-axis implies the percentage of species. The average of all the contributions of each element in all the factors is normalized, so that its sum is 1. In this study, identification of sources was conducted using major tracers based on source emission profiles from the literature. Three sources were resolved and apportioned. The first factor featured Ti (79%), Sc (78%), Ca (78%), Fe (75%), Mn (62%) and K (33%) and apportioned 32% of the total soil mass. Fe, Ca, Mn, Sc, K, Ti and Ca are common crustal elements that form the main composition of most terrestrial materials (Odukoya,

2015; Sam et al., 2015). Crustal elements are derived from parent rock materials (He et al., 2015) during weathering and erosion activities. Construction, demolition and concrete wastes could also add to the crustal elements, especially Ca, which is the principal

constituents of gypsum used during construction activities (Ingrid et al., 2014). The combination of the loaded elements indicated characteristic signatures of soil composition. This source is associated with crustal sources.

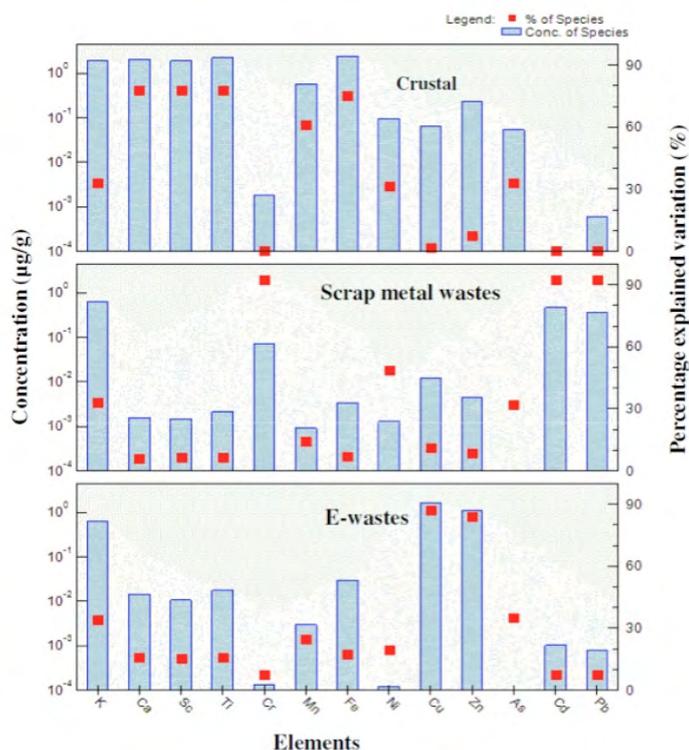


Figure 5. PMF sources resolved profile.

The second factor has high concentrations of Cr (93%), Cd (92%), Pb (92%) and Ni (48%) represents 40% of the apportioned mass. The major sources of Cr are from disposal of metal plating, chrome pigment and hardener in plastics. Ni is commonly used as part of dry cells (batteries). Tariq et al. (2013) considered Ni as one of the metals component of the several utensils used in the hospitals; and utensils in the waste stream might be a source of Ni in the soil around the dumpsites (Olabanji et al., 2015). Also, a considerable portion of Pb is from anthropogenic sources as it is an important additive in several products. It is found in automobile batteries, used in dry cells and scrap battery and in the solder of discarded printed circuit boards. Metals such as Pb and Ni are extensively used in producing surgical instruments (Tariq et al., 2013). These metals-containing instruments constitute parts of the solid wastes that finally end up at the dumpsite. This factor is identified as scrap metals wastes.

The third factor is dominated by Cu (87%) and Zn

(84%) and 28% as its constituents. Copper could be released into the soil from a high volume of irreparable electronic items and electrical accessories that are disposed of in the dumpsites. Since there is large scale use of electronic items like computers, photocopy machines at both OAU and OAUTHC, Cu might have originated from faulty or obsolete electronic items related sources. Electronic wastes are rich in Cu and it could be traced to the Cu loading in this factor (Odukoya, 2015). Robinson (2009) reported Zn to be a part of elemental constituents of doping material to enhance electrical conductance of Si in the electric circuit board of most electronic items. Hence, this factor is assigned electronic waste.

Conclusions

In this study, the elemental concentrations (K, Ca, Sc, Ti, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd and Pb) of soil samples impacted by the municipal and medical wastes

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were determined by employing PIXE spectroscopic technique. The Cf values of Cr, Mn, Fe, Ni, Cu, Zn, As, Cd and Pb were >6, classifying the soil as very high contamination. The overall Cdeg values are 176.9 and 170.4 for municipal and medical dumpsites. The PLI values of the measured elements for both dumpsites were >5, showed a progressive deterioration of the soil of the dumpsites. Based on Igeo values, the soil samples were extremely polluted with respect to Zn (5.1). All the contamination indices showed that the soils of the two dumpsite were moderately to highly polluted. The three identified factors were crustal, scrap metals waste and E-wastes. These results make it clear that there is an urgent need for the development of proper waste management strategies to safe guard the ecology and reduce the human health risks arising from the local waste stream.

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