EVALUATION OF RADIATION EMISSION AND ELEMENTAL ANALYSIS IN E-WASTE DUMPSITES

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Abstract: In this study, detailed investigationof the elemental composition occurence and concentration of selected natural radioactive nuclides in soils from e-waste dumpsite of Alaba market, Lagos state, Nigeria was carried out. The study objective is to assess the impact of e-waste management on the elemental and radiation properties of the soil. Three major e-waste dumpsites within the study area were assessed using a combination of Gamma ray spectroscopy and energy dispersive X-Ray Fluorescence. The mean specific activity of the concentration values obatained for K-40, U-238 and Th-232 were 879 ± 5, 21 ± 2 and 59 ± 7 Bq/Kg, respectively. With the exception of U-238, these concentrations were higher than the world average values. Annual effective dose and external hazard index were used to assess potential hazards which may arise from the dumpsites soil. The mean concentration of K, Ca, Cr, Mn, Fe, Ni, Cu, Zn, As, Ti, Ga, Sr and Se were 2882, 2418, 3, 332, 3259, 98, 91, 527, 59, 2399, 31, 196 and 64 mg/L, respectively. The concentration of Zn, As, Ni, and Cu were higher than the permissible limits in soil. The level of radiation dose is not likely to lead to any health hazard; nevertheless, the presence of heavy metals in the sites renders the soil unsuitable for agricultural purposes.

Keywords: E-waste, dumpsite, natural radioactivity, heavy metals.

Introduction

Advances in the electrical and electronic industries lead to the generation of electronic waste (e-waste). Lifespan of electronic products are shortened as a result of advancement in designs, compatibility, electronic, applications and marketing (Kiddee *et. al*., 2013). Currently, it is difficult to accurately determine the volume of (Waste Electrical and Electronic Equipment) WEEE globally (Bushehri *et al*., 2010; Yia-Mella *et al*., 2014). The generation of WEEE is fast growing with a fraction of about 8% of all urban waste. Managing WEEE (e-waste) is a serious global concern (Kiddee *et al*., 2013; Yia-Mella *et al*., 2014). Assessment of waste generated from computer in United States between 1997-2007 revealed 500 million while in Japan, 610 million computers were discarded (Bushehri *et al*., 2010; Kiddee *et al*., 2013). In China, five million computers are condemned as waste annually (Bushehri *et al*., 2010; Kiddee *et al*., 2013).

Effective management has failed majorly as a result of poor legislation and rapid technological advancement (Kiddee *et al*., 2013; Yia-Mella *et al*., 2014). Usually e-waste management involve recycling and disposal to landfill (Puckett *et al*.,2002; Wong *et. al*.,2007; Robinson 2009; Leung *et. al*., 2010; Kiddee *et. al*., 2013) which approaches pose serious risk to human and environment (Kiddee *et al*., 2013). For instance, heavy metals were found in air, dust, soil, sediment and fresh water around e-waste recycling sites in China (Cheng *et al*., 2003; Leung *et al*., 2010). Similar incidence was also reported in Bangalore India around recycling site (Ha *et al*., 2009). Also, landfills containing e-waste have been reported to contaminate ground water (Kiddee *et al*., 2013). Toxicity Characteristic Leaching Procedure test classified e-waste as hazardous waste (Kiddee *et al*., 2013). Due to ineffective management, large volumes of e-waste are exported to Africa.

Nigeria has the largest market for e-waste in Africa, popularly called "second hand electronics (Onwaghara *et al*., 2010; Osibanjo, 2010). After the removal of valuable parts, unused accessories are assembled and burnt. Since there is no formal recycling programme in Nigeria (Osibanjo, 2010), the predominant methods of disposal of WEEE are burning and landfilling. Burning and landfilling hazardous substances could be harmful to human and environment since the constituents of e-wastes have been reported to contain lead, mercury, cadmium and persistent organic pollutants most of which are very toxic to human health (Puckett *et al*., 2002; Wong *et al*., 2007; Robinson, 2009; Leung *et al*., 2010; Kiddee *et al*., 2013; Osibanjo, 2010). The toxic chemicals are released into the environment through burning and leaching (Balakrishnan *et al*., 2007; Olukoya 2010; Osibanjo, 2010). During the process of burning, soil, groundwater and air are contaminated (Frank, 1999; Balakrishnan *et al*., 2007; Olukoya 2010; Osibanjo 2010; Ademola *et. al*., 2014).

The environment could therefore expose to radiation from ashes and smoke fumes from dumpsites causing high risks to human health and the environment. For instance, several studies in Nigeria (Fasasi *et al*., 2003; Obed *et al*., 2005) have measured the radioactivity concentration of terrestrial radionuclides present in soil to ascertain the levels of contamination. It was reported that Nigerian foods, soil, water and particulate comprise trace amount of radionuclide and refuse dumpsites were identified as a liable recipient in containment of radioactive materials (UNSCEAR, 1993; Fasasi *et al*., 2003; Obed *et al*., 2005). This could be due to agricultural activities near dumpsites particularly horticulture farming. Accurate measurement of elemental composition and radionuclide levels in soil sample from these dumpsites will provide information which will be useful in estimating average radiation dose and metal pollution hazard in the environment (Robinson, 2009). Human exposure to telluric radiation have been reported to originate from the upper 30 cm of soil (Robinson, 2009) and only radionuclides (Th-232, U-238 and K-40) whose corresponding decay product exists in terrestrial material are of great interest. Therefore, to assess health risks to humans, estimating the distribution of radiation dose is very important (Fasasi *et. al*., 2003; Obed *et al*., 2005).

This work is focused on determination of heavy metals concentration, specific radiouclides activity and their resulting dose in top soil of WEEE dumpsites.

Materials and Methods

Sample collection

Soils from three major e-waste dumpsites (where there is current e-waste burning activities) in Alaba International Electronics Market, Ojo Local Government Area, Lagos State, Nigeria were sampled. 50-100 g of samples were collected randomly at a depth of 15 cm from each dumpsite using auger. WEEE on dumpsite ready for burning is shown in Fig. 1. A control soil sample was collected at a depth of 50 Km on the dump site. The samples were air dried at room temperature for about 2 weeks, thorougly mixed and pulverized.

Radionuclides analysis: the samples were sealed air tight in a plastic container for 28 days in-order to achieve secular equilibrium before being counted using gamma-ray spectrometer fitted with a sodium iodide detector system (IAEA 1989) available at Centre for Energy Research and Development (CERD), Obafemi Awolowo University, Ile-Ife. In order to relate effect of activity level of radionuclides in soil from dumpsites on bio-system, some important radiological hazard indices such as absorbed dose rate, annual effective dose and external hazard index were calculated.

The outdoor air-absorbed dose rates due to terrestrial gamma rays at 1 m above the ground was calculated using the relationship derived by Beck et al. (1972) which is given as;

$D = 0.042\ Ac\ (K) + 0.429\ Ac\ (U) + 0.666Ac\ (Th)$ ……. (1.0)

The annual effective doses are calculated (ICRP, 1988);

Annual Effective Dose (De) = D x Conversion factor x 1 x 365days x 24 h x 10-6 (mSv/yr) …… (2.0)

External Hazard Index (Hex) is defined as;

$Hex = A_U/370 + A_{Th}/259 + A_K/4810$ ……. (3.0)

Where: A_U; A_{Th}; and A_K are the activity concentrations of 238U, 232Th and 40K in Bqkg-1, respectively.

Fig. 1: WEEE on dumpsite at Alaba Market, Lagos

Elemental analysis: about 300 mg of the pulverized samples were made into pellets (13 mm) using the CARVER model manual pelletizing machine at a pressure of 6-8 torr. The pelletized samples were energy analyzed via the X-ray fluorescence spectrocopy facility at a voltage of 25 KeV and current of 50 µA for 1000 counts or approximately 18 minutes. Characteristic x-ray of the samples were detected by the solid state Si-detector system and spectrum acquisition was done using ADMCA software. The spectrum analysis was done using the ADMCA plus Fundamental Parameter (FP-CROSS) Software which translates the peak areas into concentration values. The elemental concentrations of the analyzed samples were statistically analyzed to show linear relationship between metals.

Statistical analysis

Statistical Package for Social Scientist (SPSS) was used for statistical analysis and correlation was considered significant at 0.05 level (2-tailed). The normally distributed data were further processed using Pearson correlation matrix.

RESULTS AND DISCUSSION

Radiological impact

The results of the gamma-ray spectrometric analyses are presented in Table 1. The radionuclides observed belong to the decay series of naturally occurring radionuclides of ^{238}U and ^{232}Th as well as the non-series ^{40}K, which accounts for largest contribution of the radionuclide present. The activity concentrations due to these three radionuclides vary from 11 ± 1 Bq/kg to 951 ± 7 Bq/kg with a mean value of 320 ± 5 Bq/kg. Specific activity concentrations for ^{238}U varied from 11 ± 1Bqkg^{-1} to 38 ± 2 Bqkg^{-1} with a mean value of 21 ± 2 Bqkg^{-1}. ^{232}Th: 37 ±11 Bqkg^{-1} – 93 ± 22 Bqkg^{-1}, mean value of 58±7 Bqkg^{-1}; and ^{40}K: 758 ±6 Bqkg^{-1} to 951 ± 7 Bqkg^{-1}, mean value of 879 ± 5 Bqkg^{-1}. From the data obtained, it is clear that the activity concentration of ^{40}K in all the samples were higher than the activity concentration of ^{232}Th and ^{238}U (Fasasi *et al*., 2003). With the exception of ^{238}U, the mean concentrations of the identified radionuclides in the samples were above world average. Table 2 lists the air absorbed dose rate, external hazard index Hex and annual effective dose for soil in the study area. The absorbed dose rate calculated for this study ranged from 24.87-62.18 nGyh-1 with a mean value of 39.04 nGyh-1 which is lower than the world average of 60 nGyh^{-1} (Tchokossa *et al*., 1999).

Table 1: Radioactivity concentrations of radionuclides in the soils of e-waste dumpsites around Alaba Market (Bqkg^{-1})

	K-40 (Bqkg^{-1})	U-238 (Bqkg^{-1})	Th-232 (Bqkg^{-1})
Site I	927 ±1	38±2	93±22
Site II	951±7	14±2	45 ±13
Site III	759±6	11±1	37±11
Mean	879±5	21 ±2	59±7
Control	317±14	19 ±14	13±3
UNSCEAR (1993)	400	35	30

Table 2: Absorbed dose rate (nGyhr^{-1}), effective dose rate (mSvyr^{-1}), and external radiation hazard (hex) index in soil of e-waste dumpsites around Alaba market.

	D (nGyh^{-1})	D_e (nSvh^{-1})	HEX
Site I	62	0.4	0.2
Site II	30	0.2	0.2
Site III	25	0.2	0.2
Mean	39	0.2	0.2
Control	30	0.2	0.2
ICRP (1991)	-	1.0	< 1.0
UNSCEAR (1993)	60	-	-

Annual effective dose (D)

To estimate the annual effective dose, account must be taken of the conversion coefficient from absorbed dose in air to effective dose and the indoor occupancy factor. Using the dose rate data obtained from the concentration

values of natural radionuclides in soil, adopting the conversion factor of 0.7 Sv/Gy absorbed dose in air to effective dose received by adults and considering that people on the average, spent 20% of their time outdoors. The annual effective dose ranges from 0.15 – 0.38 nSvh-1. The highest value in this study is lower than the world average of 1 nSvh-1. (ref ICRP, 2007). The value of this index must be less than unity in order to keep the radiation hazard to be insignificant. The internal hazard index ranged from 0.16 – 0.19 with a mean of 0.18 which is less than the threshold value set at 1 for the general public [10]. The external radiation hazard index is less than 1, which means it is safe for human to carry out their activities in the area.

Elemental composition of the soil samples

Heavy metals occur naturally in soils as a result of the weathering of the parent rock and anthropogenic activity has resulted in the widespread atmospheric deposition of these metals. Table 3 present the summary of the EDXRF results. Comparison with the average elemenetal composition with control site; concentration of Ca, Fe, Ni, Zn, Ti and Sr at the dumpsites were higher. These metals may be contained in accumulators and batteries (Ni, Zn), Cathode ray tubes (Sr), pigment (Ti, Ni) and various other electronic materials.

In all the sites considered in the study, Ga and Se were lower in concentration than in the control site, this might be due to geological composition of the site. Site II and III gave higher concentration of Fe, Ni, Ti, Sr and K, Mn, Ti, Zn respectively relative to the control site. Also, site I contains Ca, Ni, Zn, Ti and Sr at higher concentration were higher than control site I. Generally, all sites contain Ni and Zn while Se, As and Cu is predominate in at least two sites. This implies that predominant scraps burnt may be printed wiring/circuit board, accumulators, batteries, cathode ray tubes, screen coating, getter, switch, light emitting diode and solar cells. WEEE was dumped on the sites indiscriminately, though all dumpsites contain WEEE, nevertheless type of waste differs. Variation in constituent WEEE on each dumpsite could account for spatial variation in metals concentration.

According to European Union Soil Standards, the permissible concentration of Cu, Zn, Ni and Cr are 140 µg/g, 300 µg/g, 75 µg/g and 150 µg/g respectively (EU, 2002). The zinc and nickel concentration in all the sites exceeded this limit while As and Cu exceeded in two sites. Zn, Ni, As and Cu are notable among most commonly found heavy metal contaminants (Wuana et. al., 2011). The presence of these elements beyond the standard limit in soil could be regarded as toxic to human and animal (Maslin *et al*., 2000; McLaughlin *et al.*, 2000a; McLaughlin *et al.*, 2000b; Jarup 2003; Ling *et al*., 2007; Wuana *et al*., 2011). For instance, Zn in high concentration pollutes the underground water which is then leached to rivers and streams. Aquatic organisms accumulate this metal in their bodies in contaminated waterways. Human beings are exposed to risk during consumption of polluted aquatic animals, water and plants (Wuana *et al.,* 2011). Besides, environment is also adversely affected. For instance, organic matter breakdown is retarded due to interruption of microbes and annelida activities (Wuana *et al.*, 2011). Furthermore, comparison of elemental concentrations of soils around e-wastes dumpsites in Nigeria with those reported for India and China revealed Cr, Cu, Zn and Mn were common in all the dumpsites (Ha *et al.*, 2009; Kiddee *et al.*, 2013). Although the concentrations of detected elements in Nigeria dumpsites were relatively lower compared to other dumpsites, this may due to heavy recyling activities in China and India (Wong *et al*., 2007; Ha *et al.*, 2009; Leung *et al*., 2010; Onwaghara *et al*., 2010).

Table 3: Elemental concentrations in soils of e-waste dumpsites around Alaba Market

Parameters (mg/l)	Site I	Site II	Site III	Mean	Control
(K)	598±47	1745±81	6304±153	2882±94	4598±134
(Ca)	7249±162	1±1	6±1	2418±54	2333±95
(Cr)	1±1	4±1	3±1	3±1	-
(Mn)	252±9	224±9	519±14	332±11	353±13
(Fe)	1±1	9773±68	3±1	3259±23	1±0
(Ni)	78±3	124±4	93±3	98±3	9±1
(Cu)	96±2	8±1	170±2	91±1	-
(Zn)	481±9	245±7	856±12	527±9	355±12
(As)	77±2	13±1	88±2	59±2	-
(Ti)	761±34	2806±67	3631±79	2399±60	647±31
(Ga)	13.±1	48±3	31±2	31±2	96±6
(Sr)	200±26	202±26	187±24	196±25	174±18
(Se)	63.±3	73±3	55±2	64±3	253.±9

Table 4: Correlation analysis of the elemental analysis results in the waste dumpsites

	K	Ca	Ti	Fe	Cr	Mn	Ni	Cu	Zn	Ga	As	Rb	Sr	Se	Zr
K	1.00														
Ca	**0.99**	1.00													
Ti	**0.84**	**0.78**	1.00												
Fe	**0.97**	**0.99**	0.69	1.00											
Cr	0.38	0.28	**0.82**	0.14	1.00										
Mn	**0.96**	**0.98**	0.66	**0.98**	0.10	1.00									
Ni	0.00	-0.11	0.54	-0.24	**0.93**	-0.28	1.00								
Cu	**0.72**	**0.79**	0.23	**0.86**	-0.37	**0.88**	-0.70	1.00							
Zn	**0.83**	**0.89**	0.40	**0.94**	-0.20	**0.95**	-0.56	**0.98**	1.00						
Ga	0.21	0.11	0.70	-0.03	**0.98**	-0.07	**0.98**	-0.53	-0.37	1.00					
As	0.45	0.54	-0.10	0.65	-0.66	**0.68**	-0.90	**0.94**	**0.87**	-0.78	1.00				
Rb	**0.95**	**0.91**	**0.97**	**0.84**	**0.65**	**0.82**	0.32	0.46	**0.61**	0.51	0.14	1.00			
Sr	-0.95	-0.98	-0.63	-1.00	-0.07	-1.00	0.32	-0.90	-0.96	0.11	-0.71	-0.80	1.00		
Se	-0.62	-0.71	-0.11	-0.80	0.49	-0.82	**0.78**	-0.99	-0.95	**0.63**	-0.98	-0.34	**0.84**	1.00	
Zr	-0.49	-0.58	0.05	-0.69	**0.62**	-0.72	**0.87**	-0.96	-0.89	**0.75**	-1.00	-0.19	**0.74**	**0.99**	1.00

(r^2 =0.05, two-tailed)

Correlation analysis

The metals K to Zr (Table 4) displayed very high correlations. Certain metals are positively correlated while others are negatively correlated. To ascertain which of the correlations are significant in the statistic, critical multiple correlation coefficient '*r*' was obtained using the Table of significant values. Critical value '*r*' with n=5, α =0.05 is 0.754. From Table 4, K has strong positive correlation with Ca, Ti, Fe, Mn, Cu, Zn and Rb corresponding to r = 0.99, 0.84, 0.97, 0.96, 0.72, 0.83 and 0.95 respectively. Calcium an alkaline earth metal displayed strong positive correlations values (r = 0.78, 0.99, 0.98, 0.79, 0.89, 0.91), respectively with Ti, Fe, Mn, Cu, Zn and Rb. Titanium showed strong positive correlation values (r = 0.82 and 0.97) with Cr and Rb respectively. Fe displayed very strong correlation values (r = 0.98, 0.86, 0.94 and 0.84) with Mn, Cu, Zn and Rb. It was also observed that Zr and Se showed a strong correlation value of 0.99. Interestingly, some metals displayed very strong negative correlations with each other. For example, K has values of -0.95 and -0.62 with Sr and Se, respectively while Zn strongly correlated negatively with Sr (r = -0.96); Se (r = -0.950 and Zr (r = -0.99). The high positive correlation suggests strong affinity among the metals and may indicate possible common sources for the affected elements while the strong negative correlation indicates uncommon source origins for the affected elements likely to be associated with other pollution sources such as industrial and vehicular emissions.

Conclusion

Radiological activity of e-waste dumpsites does not pose any danger currently. The result of the study indicated that the mean activity concentrations of the radionuclides in the soil samples were currently within the permissible threshold limit. However, presence of Zn, As, Ni, and Cu beyond permissible limit, poses great danger to human particularly those who have direct contact. The presence of heavy metals in the sites renders the soil unsuitable for agricultural purposes. Continuous burning by the informal sector will lead to increase in the concentration of these toxic heavy metals. Relevant authorities and manufacturers of electrical and electronic components must invest on remediation of sites exposed to this waste to reduce pollution effect and penetration into food chain.

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