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Pollution sources identification, health, and radiological risk assessment of naturally occurring radioisotopes and heavy metals in waste dumpsites in Ijebu-Ode, Ogun State, Southwest Nigeria

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DOI: 10.1080/15275922.2021.2006365

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Document Version Peer reviewed version

Citation for published version (Harvard):

Gbadamosi, MR, Abayomi, AA, Afolabi, TA, Adegboye, MA, Bakare, HO, Banjoko, OO, Ogunneye, AL, Ugbomeh, IL, Jegede, DO, Ajetunmobi, AE & Bakare, TE 2021, 'Pollution sources identification, health, and radiological risk assessment of naturally occurring radioisotopes and heavy metals in waste dumpsites in Ijebu-Ode, Ogun State, Southwest Nigeria', *Environmental Forensics*, pp. 1-18. https://doi.org/10.1080/15275922.2021.2006365

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1	Pollution sources identification, health, and radiological risk assessment of naturally occurring
2	radioisotopes and heavy metals in waste dumpsites in Ijebu-Ode, Ogun State, Southwest Nigeria
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15	Abstract
16	This study evaluates human health, pollution, and radiological risk assessment of potentially toxic
17	metals (PTMs) (Pb, Cd, Cr, Ni and Zn), radioisotopes (238U, 232Th and 40K) and its associated
18	radiological indices from dumpsite soils in Ogun State, Nigeria, using a calibrated atomic absorption
19	spectrophotometer (AAS) and highly shielded γ -ray spectrometry using NaI(TI) detector. Fourier
20	transform infrared spectrometer (FTIR) complemented by X-ray diffraction (XRD) techniques were

used to evaluate the mineralogical composition of the soils. Multivariate statistical analysis was used 21 to apportion the source of PTMs and the radioisotopes. The mean concentration of Pb, Cd, Cr, Ni and 22 Zn obtained were 22.35, 17.95, 20.83, 19.02 and 75.88 mg kg-1 respectively. The activity 23 concentrations of 238 U, 232 Th and 40 K ranged from 49.71 ± 16.3 - 314.15 ± 55.2, BDL - 87.54 ± 7.38 24 and BDL - 3721.3 ± 231.6 Bqkg⁻¹respectively. The values obtained were above the global average 25 value in most of the samples. According to the four-pollution and ecological risk assessment model, 26 the dumpsite soils is strongly to low polluted and enriched with toxic metals in the order Zn > Cr >27 Pb > Ni > Cd. The estimated carcinogenic risk of the three carcinogenic PTMs for children and adults 28 were higher than the acceptable limit (1 x 10⁻⁶). The results of the PCA and HCA results are consistent 29 with the correlation coefficient analysis which showed that mixed natural, anthropogenic and 30 31 lithogenic sources mainly from aggregation of lead-containing materials in the waste on the dumpsite, vehicular emission and industrial discharges are the main sources of Pb, Ni and Cd in the three studied 32 dumpsites. Thus, dumpsite soil poses great threat to health, increased pollution and enhances the 33 radiological risk to the general population via human multiple exposure routes. 34

Keywords: Toxic metals; Radiological risk; Geo-accumulation; Mineralogical and Carcinogenic risk

37 1. Introduction

The steady increase in the volume of waste generated, coupled with the inefficient waste disposal and recycling processes in Nigeria, have continued to exacerbate the risk of potential toxic metals (PTMs) and radioisotopes contamination of the soil and water bodies. This has led to increased exposure of abiotic and biotic components of the nation's ecological systems to possible degradation, and several debilitating challenges posed to the well-being of city residents (Njoroge, 2007). Major cities in Nigeria struggle to manage physical waste efficiently while chemical and other hazardous wastes and

their impacts do not get deserved attention (Adevemi, 2011; Ikporupko, 2018). Possible hazards from 44 waste dumpsites are not only in terms of the obnoxious poisonous gases, odour and presence of 45 pathogens being released, but can also include radiation emanating from the sites (Gbadamosi *et al.*, 46 2017; Ojoawo et al., 2011). Soil is the main receiver of solid wastes and thus acts as a sink for 47 radioisotopes, potential toxic metals (PTMs) and other emerging organic and inorganic pollutants of 48 environmental and health concerns which are eventually embedded into the soil (Nyles and Ray, 49 50 1999). Several studies have shown that most of the synthetic and electronic waste (e-waste) that are found in open dumpsites contain toxic elements that are harmful to humans, plants and animals (Jibiri 51 52 et al., 2014). Plant absorption of these pollutants and their incorporation into the food chain is one of many indices used for calculating exposures frequency and performing human health risk assessment 53 (Zach, 1982; Wadey et al., 1991). Radioisotopes, potentially toxic metals (PTMs) and other emerging 54 organic and inorganic contaminants in foods from impacted soil can damage human health through 55 various absorption and biochemical pathways (Mamut et al., 2017). Long-term exposures to 56 carcinogenic metals and radioisotopes through multiple exposure routes could lead to serious health 57 effects such as chronic lung diseases, acute leucopoenia, anaemia and necrosis of the mouth 58 (Ramasamy et al., 2013). ²³²Th exposure can cause lung pancrease, hepatic, bone and kidney cancers 59 60 and leukaemia (Taskin et al., 2009).

Ijebu-Ode metropolis, being a densely populated city in Nigeria, generates a large amount of electronic and non-electronic solid waste materials which are disposed-off in open sites without any soil protection measures (Solaja et al., 2017). The solid wastes end up interacting with the soil particles, thereby changing the soil physicochemical properties (Piccolo and Mbagwu, 1997). The increased volume of wastes generated in most Nigerian urban settlements necessitates the need to correctly assess the risk posed by these wastes, suggest effective waste disposal legislation and

institute possible mitigating measures to arrest likely impacts on human health, plants, and animals. 67 Therefore, the objectives of the present study include (i) determination of the activity concentrations 68 and spatial distribution of waste enhanced naturally occurring radioisotopes materials (WENORM) 69 ²³⁸U, ²³²Th and ⁴⁰K in the representative soil samples from the selected dumpsite in Ijebu-Ode, 70 71 Southwest Nigeria (ii) to evaluate the pollution and ecological risk levels based on the waste (iii) to 72 evaluate the carcinogenic and non-carcinogenic risk of identified PTMs, radioisotopes and associated 73 radiological parameters via multiple exposure pathways in the soil samples from the dumping sites (iv) identify and apportion the main source of radioisotopes and heavy metals pollution and (v) 74 establish the relationship between the PTMs radioisotopes and their associated radiological and health 75 risk parameters. Thus, this study will facilitate the establishment of a reliable baseline data for the 76 heavy metals, radioisotopes, and its associated parameters in a typical dumpsite in Southwestern 77 Nigeria. 78

79

80 2. Materials and Methods

81 **2.1**

2.1 Description of the study area

Ijebu-ode is a densely populated town located within latitude 6°45'0' N and 6°50" North of the equator 82 and longitude 3°50" E and 4°00" East of the Greenwich Meridian (Figure 1). It is situated at 110 km 83 north-east of Lagos and within 100 km radius of the Atlantic Ocean from the Eastern fringes of Ogun 84 85 State (Nigeria) and Dahomey basin (Benin Republic). The Dahomey basin envelopes a substantial part 86 of the continental margin of Gulf of Guinea, stretching from Volta-delta in Ghana (West) to the Okitipupa ridge in Nigeria (East) (Nton et al., 2009). Dahomey basin is much of a geological interest due to 87 the reported discovery of valuable mineral deposits within the region (Nton, 2009). Three major 88 wastelands/dumpsites within the city were used for the study. These study locations were Ikoto 89

90 dumpsite (IAS) along Benin-Sagamu Expressway, Ikangba Estate dumpsite (IBS) and Olisa dumpsite



91 (ICS) respectively.



- 93 Fig. 1: Map of the Study area showing the Sampling locations
- 94 2.2 Soil sample collection and preparation

The study areas Ikoto, Ikangba and Olisa dumpsites are located within the latitude $03^{\circ}52'890''N$ and 03°56'360'' North of the Equator and longitude 06°47'169''E and 06°48'876'' East of the Greenwich Meridian. A total of sixty (60) soil samples were obtained from the three dumpsites between May-June 2018 and July 2019 at 0 – 10 cm depth, using a hand driven soil auger. Two waste free control samples within the same geological formation with the dumpsites were also sampled. In each location, the dumpsite was divided into two sections and twenty sub-samples were collected and composited

from each section to obtain representative 10 samples for the top and sub-soil respectively using 101 coning and quartering technique. Thus, twenty representative soil samples were obtained from each 102 dumpsite and three control soil samples without any anthropogenic input were also collected. The soil 103 samples were placed in a new labeled polythene bags and immediately taken to the laboratory. Their 104 respective sample locations were recorded using a hand-held Global Positioning System (GPS) 105 (Model GARMIN GPS-13). Samples were air-dried separately within a controlled chamber at room 106 107 temperature for a month to avoid further atmospheric deposition and cross contamination, and then pulverized before made to pass through 60 mesh sieves after removing the stones and debris. 108

109 2.2.1 Potential toxic metals (PTMs) Digestion Procedure and Radioisotope measurements

For the PTMs determination, 1 g of soil samples were digested and spiked with a standard solution of 110 the five metals for about 1 hour in a ternary mixture of 3:2:1 nitric acid (HNO₃), perchloric acid 111 (HClO₄) and Hydroflouric acid (HF) respectively, as described by (Gbadamosi et al., 2018a; Liu et 112 113 al., 2013). The concentrations of the PTMs were then determined using Buck Scientific (Model 210 114 VGP) Atomic Absorption Spectrophotometer (AAS). The results obtained were compared with those of the un-spiked dumpsite soil samples. The percentage recovery of the spiked soil samples were 89.3 115 % for Pb, 92.4 % for Cd, 97.4 % for Cr, 91.2 % for Ni and 86.5 % for Zn, respectively. The limits of 116 detection (LOD) were 0.08, 0.01, 0.04, 0.05 and 0.15 for Pb, Ni, Cd, Cr, and Zn and the limits of 117 quantification (LOQ) were 0.26, 0.03, 0.13, 0.17 and 0.50 mg/kg for Pb, Ni, Cd, Cr, and Zn 118 respectively. For the PTMs determination, the absorbance value for blank and working standard 119 solution were also measured. 120

For radioisotopes measurement of the dumpsite soil samples, two hundred grams (200 g) of each prepared soil samples were packed into cleaned and labeled cylindrical glass vessels and then placed in transparent polyethylene bags and sealed. These samples were safely conveyed to National Institute of Radiation Protection and Research (NIRPR), University of Ibadan, Ibadan, Southwest Nigeria. At the laboratory, the plastics were hermitically sealed with sticky tape and kept for 40 days to ensure that the parent and daughter nuclides in the sample were at secular equilibrium between radium and its gaseous decay progenies. At the end of this equilibrated period, the samples were subjected to gamma-ray spectrometer.

129 2.2.2 Quality control and statistical analysis

130 For quality assurance and quality control (QA/QC), reagent blanks and sample duplicates were incorporated in the analysis to detect any contamination in the analytical materials and to assess 131 precision and bias in the method of analysis. The recoveries ranged from 86.5 % to 97.4 %. Samples 132 were carefully conveyed to the laboratory in a sealed compartment. All glassware, sample collection 133 134 materials and digestion vessels were pre-washed with soap less detergent, soaked with dil. HNO3 135 (1+1) and then thoroughly rinsed in tandem with double distilled and deionized water. All the reagents used were of high purity analytical grade. Univariate and multivariate statistical analyses were used 136 for the analysis of radioisotopes and PTMs in the soil samples. The univariate analysis includes, mean, 137 maximum, minimum, standard deviation, coefficient of variation, standard error of the mean (SEM), 138 skewness and kurtosis. The multivariate statistical analyses (MVA) such as principal component 139 140 analysis (PCA), hierarchical cluster analysis (HCA) and correlational analysis were employed to apportion and differentiate the sources of the PTMs and radioisotopes and establish the mutual 141 influence of the variables on each other. This according to Tahri et al. (2005), will allow the 142 143 determination of the relationship between radioisotopes and their associated radiological and health parameters and PTMs in order to establish their common origin using the commercial statistics 144 software package IBM SPSS statistics (version 26, Inc., Chicago IL). MATLAB software was used 145 for the spatial distribution of the radioisotopes and PTMs in the samples. 146

147 2.3 Radioisotopes Analysis

148 2.3.1 Sample measurements

Radioisotopes 238 U, 232 Th and 40 K activity concentrations were quantified by a well calibrated γ -ray 149 spectrometry using NaI (TI) detector. The counting assembly for the natural radioisotopes 150 151 determination made up of a scintillation detector coupled to a Canberra multi-channel analyzer which consists of 7.6 mm x 7.6 mm NaI (TI) detector (Model Bicron) with adequate lead shielding which 152 reduced the background by a factor of about 0.95. The γ -ray spectrometer was calibrated appropriately 153 (energy and efficiency calibration) and tested for its linearity using the well-calibrated standard 154 gamma source outsource from the International Atomic Energy Agency (IAEA), laboratories, Vienna, 155 Austria (Ademola et al., 2008). The resolution of the NaI (TI) detector is 7.5 % at 0.662 MeV of ¹³⁷Cs. 156 Each of the samples was measured for 18 000 sec in order to obtain good statistics for the 157 radioisotopes. Also, measurements were repeated at intervals for quality control and assurance 158 purposes as well as to maintain the stability of the counting system. The background radiation due to 159 the radioisotopes in the vicinity around the detector was measured using an empty plastic container, 160 which was measured in the same manner as the soil and reference sample (Jibiri et al., 2014). The net 161 radioisotopes activity concentrations were obtained by subtracting the background spectrum from the 162 measured spectra. Three intensity regions were identified in the spectrum which corresponds to 1.460 163 MeV for (⁴⁰K) content, 1.760 MeV for (²¹⁴Bi) and ²³²Th content determined from 2.614 MeV for 164 (²⁰⁸TI) in the samples. The below detectable limit (BDL) of each radionuclide were determined from 165 the background radiation spectrum for the same counting time as was done for the dumpsite soil 166 samples and are given as 0.037 Bq kg⁻¹ for ²³⁸U, 0.007 Bq kg⁻¹ ²³²Th and 0.18 Bq kg⁻¹ for ⁴⁰K 167 168 respectively.

169 2.4 Radiological and health hazard parameters

In order to evaluate the γ -ray radiation and health hazards impact of the radioisotopes ²³⁸U, ²³²Th and ⁴⁰K in the dumpsites and consider the radiological hazards of the soil to humans, suitability of the soils for building constructions, farming, remediation and other purposes. Even though total activity concentrations of radioisotopes were determined, it does not provide the exact indication of the total radiation hazard. Therefore, a comprehensive estimation of these radiological and health hazard parameters to humans become necessary. Table S1 gives details formulas for each radiological and health hazard index evaluated in this present study.

177 2.5 Assessment of Potentially toxic metals Pollution and Ecological Risk

Several approaches have been used to evaluate the pollution, contamination and ecological risk of PTMs in the soil. In this present study, four of these indices such as index of geo-accumulation (I_{geo}), contamination factor (CF), pollution loading index (PLI) and potential ecological risk index (PERI) were extensively used to evaluate the pollution and ecological risk of the soil to biological organisms in the environment.

183 2.5.1 The index of geo-accumulation (Igeo)

The Index of geo-accumulation (I_{geo}) was used to evaluate potentially toxic metals (PTMs) pollution levels resulting from anthropogenic activity based on the use of a single toxic metal in the soils (Men et al., 2018). The index has been widely applied in European trace metals since the late 1960 and is defined as follows:

188
$$I_{geo} = \log_2\left(\frac{c_n}{1.5 B_n}\right) \tag{1}$$

Where C_n and B_n are the levels of potentially toxic metals, n, in the soils and the geochemical background levels of the corresponding toxic metals n respectively. The constant 1.5 represents the correction factor due to anthropogenic/lithogenic effects (Tian et al., 2017). The following classifications were used for Igeo: Igeo value > 5 (extremely polluted), Igeo between 4 and 5 (heavily-extremely polluted), Igeo between 3 and 4 (strongly polluted), Igeo between 2 and 3 (moderately-strongly polluted), Igeo values between 1 and 2 (moderately polluted), Igeo between 0 and 1(unpolluted-moderately pollution), Igeo value ≤ 0 (unpolluted) (Hua et al., 2018).

196

197 2.5.2 Contamination Factor and Pollution Loading Index

Contamination factor (CF) is a vital contamination evaluation method that determined the degree of contamination of the sampling site by PTMs (El-Amier *et al.*, 2017). The CF of each metal is calculated as the ratio of individual toxic metal concentrations to the background values, and is defined by this equation (Bourliva et al., 2018; Gbadamosi et al., 2018a).

202 Contamination Factor =
$$\frac{(C_{metal})_{sample}}{(C_{metal})_{background}}$$
(2)

CF is ranking from 1-6 was used to evaluate the level of contamination based on the strength: $CF \le 0$: non-contaminated, $0 < CF \le 1$: slightly contaminated; $1 < CF \le 3$: moderately contaminated; $3 < CF \le$ 5: considerately contaminated; $5 < CF \le 6$: strongly contaminated; CF > 6: very strongly contaminated. (Gabarrón *et al.*, 2017).

207 2.5.3 Pollution loading index (PLI)

PLI allow an easy but relative means of evaluating the total pollution and quality of the soil from the selected sites (Chakravarty and Patgiri, 2009). It is mathematically defined as the nth root of the multiplication of contamination factors (CFn) expressed as follows:

211
$$PLI = (CF_1 x CF_2 x CF_3 x CF_n)^{\frac{1}{n}}$$
 (3)

212 Where n is the number of PTMs (in the present study, n=5). PLI is a basic and convenient pollution 213 assessment method for toxic metals. The degree of pollution using PLI is classified as follows: PLI < 1 (no pollution), low pollution ($1 < PLI \le 2$), moderate pollution ($2 < PLI \le 3$), and heavy pollution (PLI > 3) (Jamal *et al.*, 2018; Gbadamosi et al., 2018a).

216

217 2.5.4 Potential ecological risk index (PERI)

PERI is a powerful technique for quantitatively express the potential ecological risk posed by individual toxic metals in the selected dumpsites soils. This method considers four pollutants characteristics such as concentration, nature, toxicity, and sensitivity of the pollutants to the water body from the soil (Gbadamosi *et al.*, 2018a; Zhang *et al.*, 2016). PERI can be determined using the formulas:

223
$$PERI = \sum_{i=1}^{n} E_{r}^{i} = \sum_{i=1}^{n} T_{r}^{i} x C_{f}^{i} = \sum_{i=1}^{n} T_{r}^{i} x \frac{C_{m}^{i}}{C_{b}^{i}}$$
(4)

Where E_r^i is the potential ecological risk factor for each toxic metal, i, T_r^i is the toxic response factor of the individual toxic metals in the dumpsite's soils. C_f^i is the contamination factor of the toxic metals, C_m^i and C_b^i are the concentrations of the toxic metals in the sample and background value of the metals, respectively. Based on literature, T_r^i for Pb, Cd, Cr, Ni and Zn are 5, 30, 2, 5 and 1, respectively (Zhang *et al.*, 2016). PERI is categorized into four as: PERI < 150 (low ecological risk), 150 ≤ PERI < 300 (moderate ecological risk), 300 ≤ PERI < 600 (high ecological risk) and PERI > 600 (very high ecological risk) (Tian *et al.*, 2017).

231

232 2.6 Human Health risk assessment

The human health risk models including the carcinogenic and non-carcinogenic risk estimation of the various organic and inorganic pollutants were developed by USEPA and have been globally adopted (USEPA, 2011; 2002). Three human exposure pathways were used to estimate the human average daily dose (ADD) of the potentially toxic metals (PTMs) via ingestion, inhalation and dermal absorption using the equation provided in Table S2. A detailed explanation of the mathematical
formulas was presented in Table S2.

The Hazard quotient (HQ) is widely used to determine the non-carcinogenic risk assessment for a single metal or hazard index (HI) for multiple metals via multiple exposure pathways. This is calculated as the ratio of the average daily dose (ADD) and the reference dose (RfD) for a given metals.

243 HQ (dimensionless) =
$$\frac{\text{Average daily dose during exposure to PTMs (ADD)(mg kg^{-1} day^{-1})}{\text{RfD (mg kg^{-1} day^{-1})}}$$
(5)

244
$$\operatorname{HI} = \sum \operatorname{HQ}_{i} = \sum \frac{\operatorname{ADD}_{i}}{\operatorname{RfD}_{i}}$$
 (6)

HI or HQ determines the magnitude of adverse health effects. Adverse health effects are unlikely to occur if HQ or HI < 1 and the magnitude of risk increases as HI or HQ increases to a unity safe level. High chronic risk effect is indicated if HQ > 10 (USEPA, 2001; 2011; Ogunbanjo et al., 2016).

The carcinogenic risk assessments are determined by estimating the probability of individuals developing any cancer type over time because of prolonged exposure to the carcinogenic agent in the study site. The carcinogenic risk can be estimated using the equation:

251

Cancer risk = ADD x PF⁽⁷⁾

Where PF (mgkg⁻¹day⁻¹) is the potency factor which is the slope of the dose-response curve (Table 3).
The cumulative cancer risk can then be calculated from:

254 Total cancer risk (TCR) =
$$\sum_{i=1}^{n} ADD_i PF_i$$
 (8)

Where PF_i is the potency factor for the metal i (mg kg⁻¹day⁻¹) and ADD_i is the average daily dose (mg kg⁻¹ day⁻¹). The accepted tolerable risk for regulatory purposes is between 1 x 10⁻⁶ \leftrightarrow 1 x 10⁻⁴ (US EPA, 2011; Li *et al.*, 2018). This means that if the estimated cancer risk is lower than 1 x 10⁻⁶, there is

a probability of one chance in a million of an individual's developing cancer over the lifetime and this
risk are considered negligible (Li *et al.*, 2018).

260

261 2.7 Mineralogical characterization using FTIR and XRD

The mineralogical characterization of the soil was determined using Perkin Elmer RX1 Fourier 262 transform infrared spectrometer (FTIR). The IR spectra for all the samples were recorded in the region 263 4000-650 cm⁻¹. Sample preparation and procedure for mineral analysis by FTIR were clearly 264 presented in the earlier published work (Gbadamosi et al., 2018b). However, for XRD, the diffraction 265 pattern was determined by Rigaku Ultimate IV X-ray diffractometer with a curve of graphite 266 monochromator and a radiation source of CuKα. The diffraction patterns were obtained in the range of 267 20 and region between 20°-80° and with a scan speed of 1°/min. The obtained XRD patterns for the 268 samples were processed by matching the diffraction patterns with a joint committee on powder 269 270 diffraction standard (JCPDS) database (Berry, 1974).

271 **3** Results and Discussion

272 3.1 Activity concentrations of radioisotopes and radiological characterization of the dumpsites soil samples

Table 1: Statistical summary of the Activity concentration and Radiological Characterization of dumpsite soil samples (n=60) and control (n=2).

274	Arithm	netic mean	Maximum	Minimum	Standard deviation	CV (%)	SEM	Skewness k	Curtosis W	Vorld average ^a
275	Concentration of ²³⁸ U(Bqkg ⁻¹)	142.20 ± 25.9	314.15 ± 55.2	49.71±16.3	60.7	36.1	7.83	1.09	0.91	33
276	Concentration of ²³² Th(Bqkg ⁻¹)	16.11 ± 3.00	87.54 ± 7.38	BDL	17.3	90.4	2.23	2.53	8.32	45
277	Concentration of ${}^{40}K(Bqkg^{-1})$	$362.30{\pm}\ 27.6$	3721.3 ± 231.6	BDL	580	148.8	74.9	4.24	24.2	420
278	Absorbed dose rate D_R (nGyhr ⁻¹)	90.54 ± 14.9	215.95 ± 25.6	40.64 ± 10.6	35.7	33.8	4.60	1.21	2.42	57
279	Radium equivalent Raeq(Bqkg ⁻¹)	193.14±32.3	420.30±55.4	84.9±23.0	74.2	32.9	9.57	0.94	0.97	370
280	AEDE _{outdoor} (µSvyr ⁻¹)	111.1±18.3	265.02±31.4	49.88±13	43.8	33.8	5.65	1.21	2.42	70
281	AEDE _{indoor} (µSvyr ⁻¹)	444.5±73.3	1060±126	199.5±51.9	175	33.8	22.6	1.21	2.42	450
282	AGDE (µSvyr ⁻¹)	620.5±101.2	1577.5±155.6	283.7±71.4	350	34.7	32.3	1.44	3.75	1000
283	$ER(\mu Rhr^{-1})$	380.5±62.6	922.1±105.3	172±43.6	151	34.0	19.4	1.24	2.66	600
284	H _{ext}	0.52±0.10	1.14±0.15	0.23±0.06	0.20	32.9	0.03	0.94	0.96	≤ 1
285	H _{int}	0.91±0.16	1.70 ± 0.30	0.36±0.11	0.34	32.2	0.04	0.76	-0.11	≤ 1
286	Iγr	0.68±0.11	1.69±0.19	0.36±0.11	0.34	34.4	0.04	1.36	3.35	≤ 0.5
287	AUI	1.54±0.28	3.00±0.51	0.58±0.17	0.61	33.8	0.08	0.81	-0.07	≤ 2
288	ELCRx10-3outdoor	388.9±64.1	928±110	175±45.4	153	33.8	19.8	1.21	2.42	290
289 290 291 292 293 294 295 296 297 298	Control 1: 238 U (Bqkg ⁻¹) Control 1: 232 Th (Bqkg ⁻¹) Control 1: 40 K (Bqkg ⁻¹) Control 1: D _R (nGyhr ⁻¹) Control 1: Raeq (Bqkg ⁻¹) Control 2: 238 U (Bqkg ⁻¹) Control 2: 232 Th (Bqkg ⁻¹) Control 2: 40 K (Bqkg ⁻¹) Control 2: D _R (nGyhr ⁻¹) Control 2: D _R (nGyhr ⁻¹) Control 2: Raeq (Bqkg ⁻¹)	$\begin{array}{c} 24.30{\pm}11.32\\ 4.55{\pm}0.42\\ 54.20{\pm}9.55\\ 16.24{\pm}5.88\\ 34.98{\pm}12.66\\ 10.32{\pm}5.43\\ 2.14{\pm}0.21\\ 21.32{\pm}5.61\\ 6.95{\pm}2.87\\ 15.02{\pm}6.16\\ \end{array}$	- T :							

The value of the activity concentrations due to the naturally occurring radioisotopes ²³⁸U, ²³²Th and 301 40 K ranged from 49.71 ± 16.3 Bq kg⁻¹ - 314.15 ± 55.23 Bqkg⁻¹, BDL - 87.54 ± 6.75 Bqkg⁻¹ and BDL 302 - 3721.25 ± 231.57 Bq kg⁻¹ with an average value of 142.20 ± 25.9 , 16.11 ± 3.00 and 362.30 ± 27.6 303 Bq kg⁻¹respectively (Table 1). It was observed that in all the soil samples activity concentrations of 304 radioisotopes ²³⁸U were about 2 to 11 times higher than the world average value of 33Bq kg⁻¹ 305 (UNSCEAR, 2000) (Fig. 2). The average activity concentrations of ²³⁸U were about 5 times higher 306 than the world average value in the dumpsites. Activity concentrations of ²³²Th in soil samples were 307 below the world average value of 45 Bq kg⁻¹ except for soil sample ICTS5, ICTS10, ICTS2 and 308 ICTS3 with activity concentrations of 45.21 ± 3.50 Bq Kg⁻¹, 53.21 ± 5.52 Bq Kg⁻¹, 76.89 ± 7.38 Bq 309 Kg⁻¹, and 87.54 ± 6.75 Bg Kg⁻¹, which are 1-2 times higher than the world average value. The activity 310 concentrations of ⁴⁰K in all the top and sub soil samples from Ikoto dumpsite exceed the world 311 average value of 420 Bq kg⁻¹. More so, five and one soil samples from Ikangba and Olisa dumpsites 312 respectively have higher ⁴⁰K values than the recommended limit of 420 Bq Kg⁻¹ (Fig. 2). The average 313 values of the ⁴⁰K radioisotope were higher than the world average value of 420 Bg kg⁻¹ in all the 314 samples. In all sampling locations, the activity concentrations of the radioisotope were in the order: 315 40 K > 238 U > 232 Th (Fig. 2). In few of the sampling points, the activity concentrations of 40 K and 238 U 316 were extremely high; this may be attributed to the extensive use of NPK fertilizer, the solubility and 317 mobility of uranium rich rock or the large deposit and aggregation of clinical wastes containing 318 uranium in the study areas. 319





The average absorbed dose rate (D_R) due to γ -radiation inhaled in air at 1 m above the ground surface for the uniform distribution of the naturally occurring radioisotopes ²³⁸U, ²³²Th and ⁴⁰K for all samples were calculated using equation based on the guidelines provided by (UNSCEAR, 2000) (Table S1). The absorbed dose (D_R) ranged from 40.65 ± 10.6 - 215.95 ± 25.6 nGyhr⁻¹ with an average value of 90.54 ± 14.9 nGyhr⁻¹ (Table 1). This estimated average value of D_R was higher than the world average value of 57 nGyhr⁻¹ (population weighted average) (UNSCEAR, 2000) (Fig. 3).

Radium equivalent activity (Raea) introduced to define uniformity in the distribution with respect to 329 radiation exposure due to the naturally occurring radioisotopes in the soil samples (UNSCEAR, 330 2000). Ra_{eq} is the weighted sum of 238 U, 232 Th and 40 K based on the assumption that 370 Bq kg⁻¹ of 331 ²³⁸U, 259 Bqkg⁻¹ of ²³²Th and 4810 Bqkg⁻¹ of ⁴⁰K gives the same γ-radiation dose rate (Straden, 332 1979). The maximum value of Raeq must be <370 Bqkg⁻¹ for the radiological effects to be 333 insignificant on the people living or working around the dumpsites i.e., scavengers or the nomadic 334 farmers. The value of Ra_{eq} obtained ranges from $84.9 \pm 23.0 \leftrightarrow 420.30 \pm 55.4$ Bq kg⁻¹ with an 335 average of 193.14 \pm 32.3 Bqkg⁻¹ (Table 1). The values obtained for Ra_{eq} were lower than the world 336 average value of 370 Bq kg⁻¹ except at four sampling points IATS1, IASS3, ICTS10 and ICSS3 337 (Ikoto topsoil and subsoil, Olisa topsoil and subsoil), (Fig. 3). The average value for Raeq were below 338 the recommended world average of 370 Bq kg⁻¹(UNSCEAR, 2000) in all the samples and meet the 339 340 recommended limit set by (OECD, 1979). The calculated outdoor and indoor AEDE values as shown in Table 1 ranges from $49.88 \pm 13 - 265.02 \pm 31.4 \ \mu \text{Svyr}^{-1}$ and $199.5 \pm 51.9 - 1060 \pm 126 \ \mu \text{Svyr}^{-1}$ (Fig. 341 3) with an average value of 111.1 \pm 18.3 μ Svyr⁻¹ and 444.5 \pm 73.3 μ Svyr⁻¹ respectively. This 342 calculated average values for the outdoor and indoor AEDE were higher than the recommended limit 343 of 70 and 410 (UNSCEAR, 2000). These higher values for the AEDEs may be attributed to higher 344 ²³⁸U and ⁴⁰K in the soils. 345

Annual genetically significant dose equivalent (AGDE), widely used to determine the genetic significance of the yearly dose equivalent (Morsy et al., 2012) was calculated to range from 283.7 ± 71.4 - 1577 ± 155.6 μ Svyr⁻¹, with an average value of 620.5 ± 101.2 μ Svyr⁻¹ respectively. These AGDE values obtained were higher than the worldwide average value of 1000 (UNSCEAR, 2000) at four out of the sixty sampling points (6.7 %) and moderately high at five of the sampling points (8.3 %). The highest AGDE of 1577 ± 155.6 μ Svyr⁻¹ values was obtained in IASS3 (Fig. 3).



354 Fig. 3: Radiological parameters of the dumpsites soil and control samples

356 3.2 Radiological and Health Hazard Indices

The external radiation hazard index (Hext), a globally used hazard index which reflects the external 357 exposure level due to γ -radiation in any environmental samples, was calculated for the samples. The 358 values obtained ranged from 0.23 ± 0.06 to 1.14 ± 0.12 (Table 1). The average value of 0.52 ± 0.10 359 obtained for Hext in all the soil sampled was less than the acceptable limit of 1. All the samples had 360 Hext value less than unity except for samples IATS1, IASS3, ICTS10 and ICSS3 which is the Ikoto 361 top and sub-soil and Olisa top and subsoil respectively (Fig. S1). The internal hazard (H_{int}) varied 362 from 0.36 ± 0.11 to 1.70 ± 0.30 with an average value of 0.91 ± 0.16 . This value was higher than the 363 recommended value which must be ≤ 1 . Hence, these areas may pose significant radiological risks to 364 the inhabitants owing to the harmful effects of ionizing radiation emanating from the dumpsites. 365 These calculated H_{int} values were above the recommended value in 47 % of the samples (28 sampling 366 points) (Fig. S1). 367

To further evaluate the radiation hazards associated with the dumpsite soil samples, another radiation 368 hazard index known as γ -radioactivity level index (Iyr)/ γ -representative level index (RLI) was 369 370 determined. Based on (UNSCEAR, 2000) guidelines, for radiological effects to be considered negligible, the values of each H_{ext} , H_{int} must be ≤ 1 and Iyr must be ≤ 0.5 . The calculated value y-ray 371 372 hazard index of the soil samples ranged from 0.36 ± 0.08 - 1.69 ± 0.17 with an average value of 0.68 \pm 0.11, respectively. The calculated values of Iyr were higher than the recommended value of 0.5 in 373 all the sampling points except in IBTS2, IBSS1, ICSS5 and ICSS8 (Fig. S1) where the values were 374 below the recommended threshold. Therefore, caution should be taken in using the soils from these 375 dumpsites as bulk building materials because their γ -index values were higher than the upper limit set 376 by (UNSCEAR, 2000). 377

The dumpsite soil samples were also examined to evaluate whether they satisfy the dose criteria when used as a building material. For this reason, the activity utilization index (AUI) was calculated. The calculated values for AUI ranged from $0.58 \pm 0.20 - 3.00 \pm 0.36$ with an average value of 1.54 ± 0.28 respectively (Table 1). The calculated values for AUI were higher than the recommended value/worldwide average value of ≤ 2 in nineteen out of the sixty samples analyzed (31.6 % of the sampling points) and moderately high in eighteen of the samples (30 %) (Fig. S1). The average value of AUI was moderately high in the samples. This shows that the present dumpsites soil samples cannot be used for safe construction of a building or agricultural purposes without adequate soil remediation or recontamination process.

387 **3.3** Excessive Lifetime cancer risk (ELCR) and exposure rate (ER)

The outdoor excessive life cancer risk (ELCRout) was calculated on the annual outdoor effective dose 388 equivalent and the results were presented in Table 1. The outdoor excessive life cancer risk (ELCRout) 389 ranged from $175 \pm 45.5 - 928 \pm 94.6$ with an average value of 388.9 ± 64.1 respectively (Table 1). 390 ELCR_{outdoor} values calculated were higher than the recommended limit of 290 (UNSCEAR, 2000) in 391 about 88 % of the samples (Fig. S2). Consequently, the average value was higher than the world 392 average value in all the dumpsites soil samples. This result shows that the lifetime cancer risk due to 393 exposure using this dumpsite soils for 70 years maximum duration is high. Hence, the subsequent use 394 of this dumpsite soil for building construction, agriculture or for soil remediation studies and other 395 purposes should be discouraged. 396

The calculated rate of exposure (ER) of an individual and scavengers to these naturally occurring radioisotopes in the selected dumpsites was higher than the maximum limit in seven locations (IATS1, IASS1, IASS3, ICTS3, ICTS8, ICTS10 and ICSS3) and moderately high in three sampling points (ICTS6, ICTS7 and ICTS9) respectively (Fig. S2) (Table 1) (UNSCEAR, 2000). These results revealed that Ikoto and Olisa dumpsite has a higher ER value. Hence the people living near these dumpsites should be aware of the inherent effects of exposure to this harmful ionizing radiation emanating from these dumpsites on daily basis.

Table 2: Statistics summary of the potentially toxic metals (PTMs) (mg kg⁻¹) in the dumpsite soil

406	samples	(n=60),	control	(n =2)) and	guidel	ines va	lues
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PTMs	$Pb (mg kg^{-1})$	$Cd (mg kg^{-1})$	$Cr (mg kg^{-1})$	Ni $(mg kg^{-1})$	$Zn (mg kg^{-1})$
Arithmetic Mean	22.35	17.95	20.83	19.02	75.88
Max.	41.82	29.21	34.87	31.12	142.54
Min.	11.14	8.14	11.43	9.23	23.76
Range	30.68	21.07	23.44	21.89	118.78
Median	20.15	18.06	20.26	19.14	67.06
SD	5.61	4.38	5.25	3.09	37.92
RSD or CV (%)	32.7	27.5	25.1	26.0	42.7
Kurtosis	0.06	-0.48	0.02	0.24	-1.04
Skewness	0.74	-0.03	0.11	0.11	0.48
SEM	0.94	0.64	0.67	0.64	4.19
ABV	11.39	6.95	39.59	7.34	13.55
SOGs	50	0.4	-	60	300
MAL (Austria)	100	5	100	100	300
MAL (Canada)	200	8	100	100	400
MAL (Poland)	100	3	100	100	300
MAL (Japan)	400	-	-	100	250
MAL (Germany)	500	2	200	100	300
MAL (Great Britain)	100	3	50	50	300
(DEP, Australia)ª	300	3	50	60	200
(CCME, Canada) ^b	70	1.4	64	50	250

^a DEP (Department of Environmental Protection) (2003).; ^bCCME (Canadian Council of Ministers of
the Environment) (2003).

409 **3.4** Potentially toxic metals (PTMs) in the dumpsite soils

The descriptive statistics of potentially toxic metals (PTMs) concentrations in the dumpsite soil 410 samples and the corresponding background samples are presented in Table 2. The mean 411 concentrations of Pb, Cd, Cr, Ni and Zn ranged from 11.14 - 41.82, 8.14 - 29.21, 11.43 - 34.12, 9.23 412 -31.12 and 23.76 - 142.54 mg kg⁻¹ with a mean value of 22.35, 17.95, 20.83, 19.02 and 75.88 mg kg⁻¹ 413 ¹ respectively (Fig. 4). Among the five studied PTMs, Zn, Pb, and Cr had the highest mean 414 415 concentrations. Coefficient of variation which is a measure of relative dispersion within the 416 concentrations of the PTMs defined as the ratio of the standard deviation to the mean concentration of the PTMs. The greater the CV, the greater the level of dispersion around the mean. If $CV \le 20\%$, it 417 shows low dispersion; if $21\% < CV \le 50\%$, it is referring to as moderate dispersion; $50\% < CV \le$ 418 100% is regarded as high dispersion or variability; while CV value above 100% is considered as high 419 420 dispersion or variability (Karim et al., 2015). The CV of the PTMs increased in the order of Zn (42.7) > Pb (32.7) > Cd (27.5) > Ni (26.0) > Cr (25.1) (Table 2). The CV for the five PTMs in this study 421 indicated that the PTMS varied moderately with respect to the different samples. Kurtosis and 422 skewness are two main measures of the degree of asymmetry in relation to normal distribution of 423 variables (Liu et al., 2017). The PTMs were positively skewed towards lower concentrations except 424 Cd, Pb, Cr and Ni showed a heavy tailed distribution with positive kurtosis and Cd and Zn shows a 425 lightly tailed distribution with negative kurtosis. Moreover, the mean and maximum concentration of 426 Cd among the PTMs measured exceeds the maximum allowable limit (MAL) for Cd in Austria, 427 Canada, Poland, Japan, Germany and Great Britain (Table 2). 428





Fig. 4: PTMs distribution in the dumpsite soil and control samples

433 3.5 Spatial Distribution of PTMs and radioisotopes using MATLAB

The degree of activity concentrations of radioisotopes²³⁸U, ²³²Th, ⁴⁰K and potentially toxic metals 434 (PTMs) Pb, Cd, Cr, Ni and Zn in the three dumpsites are shown in Fig. 5. The concentrations are 435 presented in three-dimensional distributions using MATLAB (Miao et al 2014), where the degree of 436 concentrations indicated by the colour bar. As shown in the colour bar (Fig. 5), the trend of 437 concentrations is upward with different concentration values ranging from 0 mg/kg to the higher 438 concentration value (3500 mg/kg). The higher concentrations value of radioisotopes ²³⁸U, ²³²Th and 439 ⁴⁰K in Fig. 5 (a), (b) and (c) are indicated by yellow, purple and red respectively, while those of 440 PTMs are shown in Fig. 5 (d), (e),(f),(g), and (h) maintain the similar colour trend (pink) except the 441 442 Fig. 5 (f) with a different control colour. This is because the concentrations of Cr among the PTMs in the control samples are higher than what is obtained in the three dumpsites soils. The concentration 443 of the control ranged from 33.45 mg/kg - 45.72 mg/kg against the Cr value in the three dumpsites 444 which ranged from 11.43 mg/kg to 27.21 mg/kg (Table 2). 445



Fig. 5: Activity concentrations of radioisotopes (a) ²³⁸U, (b) ²³²Th, (c) ⁴⁰K, and PTMs (d) Pb, (e) Cd,
(f) Cr, (g) Ni, and (h) Zn in the study area (the colours in the legends represent the corresponding concentration values of radioisotopes and PTMs in dumpsite and control soils).

450 **3.6** Assessment of Pollution and Ecological Risk Indices of Potentially toxic metals (PTMs)

The potential toxic metals pollution and ecological risk assessment such as index of geoaccumulation (Igeo), contamination factor (CF), pollution loading index and potential ecological risk index (PERI) were used to evaluate the degree of PTMs pollution of the dumpsite soils (Table 3). The Igeo of the five PTMs in the dumpsites ranged from 1.93 - 2.50, 1.58 - 2.13, 2.48 - 2.96, 1.66 - 2.18 and 2.33 - 3.11 for Pb, Cd, Cr, Ni and Zn, respectively. Higher Igeo values were reported for Zn Cr and Pb (Table 3). Based on the index of geo-accumulation obtained, the dumpsite soil was moderately to strongly polluted with the PTMs. The estimated value of the contamination factor (CF)

458	revealed that the dumpsite soils was moderately to considerably contaminated with Pb (0.98 - 3.67),
459	Cr (1.00 - 3.06), Cd (0.72 - 2.57) and Ni (1.26 - 2.58), strongly contaminated with Zn (2.09 - 12.52)
460	respectively. The level of PLIs in the study sites varied from 0.40 - 5.29, indicating that selected
461	dumpsite showed low pollution for Pb, Cd, Ni and heavily polluted with Zn. The potential ecological
462	risk index (PERI) which represents the sensitivity of various microorganisms to toxic metals in the
463	area and indicate the risk of the hazardous metals in the soils (Islam et al., 2015) are as shown in
464	Table 3. The potential ecological risk index (PERI) of the five PTMs increases in the following order:
465	Cr < Zn < Ni < Pb < Cd (Table 3). The results showed that the dumpsites posed a greater ecological
466	risk through the PTMs to the biological communities in the study area.

Table 3: Statistical summary of Igeo, CF, PLI and PERI

Potentially toxic metals (PTMs)	Range of PTMs concentrations (mgkg ⁻¹)	Igeo	CF	PLI	PERI
Pb	11.14 - 41.82	1.93 - 2.50	0.98 - 3.67	≤1.9	4.90 - 18.4
Cd	8.14 - 29.21	1.58 - 2.13	0.72 - 2.57	≤1.9	21.5 - 77.0
Cr	11.43 - 34.87	2.48 - 2.96	1.00 - 3.06	≤ 0.4	2.01 - 6.13
Ni	9.23 - 31.12	1.66 - 2.18	0.81 - 2.73	≤ 1.8	4.05 - 13.7
Zn	23.76 - 142.54	2.33 - 3.11	2.09 - 12.52	≤ 5.29	2.09 - 12.5

468

469 **3.8** Potential Human risk assessment of PTMs - Carcinogenic and Non-carcinogenic risk

The human health risk assessment which was calculated based on the concentrations of PTMs in the 470 dumpsite soils are shown in Table 4 and in supplementary materials (Table S3a and S3b). The mean 471 non-carcinogenic risk (HQ) of PTMs through the three exposure pathways for children and adults in 472 the dumpsite soil are in the order of HQ_{dermal} > HQ_{ingestion} > HQ_{inhalation} respectively (Table S3b). 473 Dermal contact contributed from about 69 % of the THI with the highest obtained for Zinc. These 474 results are in consistent with previous studies (Jamal et al., 2018; Gbadamosi et al., 2018a; Zhou et 475 476 al., 2020). The carcinogenic risk (CR) for the three carcinogenic metals for children and adults were in the order: Cr (9.61E-04) > Cd (1.24E-04) > Ni (1.76E-05) and Cr (6.92E-05) > Cd (2.05E-05) > Ni 477

(3.75E-07) (Table 4). More so, the total carcinogenic risk (TCR) for children with respect to the three 478 exposure pathways is about 1220 times greater than for adults. This revealed that the children are 479 480 likely to suffer more carcinogenic risk on exposure to the PTMs in the dumpsite soils samples via multiple exposure routes. These values obtained for children and adults were higher than the 481 acceptable limit (1E-06). This showed that the dumpsite soil samples posed greater carcinogenic 482 health risk for children and adults. Overall, the non-carcinogenic risks of the five PTMs were all <1, 483 484 which shows that there would be no non-carcinogenic risk from the concomitant effect of the five 485 PTMs in the study areas.

488	Table 4: potency factors, carcinogenic and non-carcinogenic risks of the heavy metals in the dumpsite soil to children and adults								
489	PTMs	EPCsoil		Children		Adults			
490		$(mg.kg^{-1})$	PF (mg/kg-day) ⁻¹	THI	Cancer risk (CR)	THI	Cancer risk (CR)		
491	Pb	22.35		8.29E-02		1.33E-02			
492	Cd	17.95	8.40E-01	2.91E-01	1.24E-04	1.49E-01	6.92E-05		
493	Cr	20.83	6.30E+00	1.04E-01	9.61E-04	4.05E-02	2.05E-05		
494	Ni	19.02	4.20E+01	1.23E-02	1.76E-05	1.86E-03	3.75E-07		
495	Zn	75.88		2.84E-01		5.38E-01			
496	∑HI			7.74E-01		7.42E-01			
497	∑CR				1.10E-03		9.01E-05		

Table 5: The observed absorption band and their corresponding minerals identified from FTIR spectra

SI. No	Range of observed wave number (cm ⁻¹)	Sample ID	Minerals
1.	696.52 - 1059.14	IATS1 - IATS10	Quartz
	1692 - 1868.3	IATS1 - IATS10	Quartz
	669.96 - 1008.45	IASS1 - IASS10	Quartz
	753.52 - 1003.52	IBTS1 - IBTS10	Quartz
	756.60 - 1034.34	IBSS1 - IBSS10	Quartz
2.	911.39 - 1004.29	ICTS1 - ICTS10	Kaolinite
	3621.75 - 3695	ICTS1 - ICTS10	Kaolinite
	3426 - 3646.3	ICSS1 - ICSS10	Kaolinite
3.	2003.5 - 2301	IATS1 - IATS10	Organic carbon
	2859.8 - 2916.9	IATS1 - IATS10	Organic carbon
	2915.9 - 2921.6	ICTS1 - ICTS10	Organic carbon
4.	3612 - 3896	IBTS1 - IBTS10	Carbonates











523 **3.7** Mineralogical characterization (FTIR and XRD)

524 3.7.1 FTIR analysis

The soil samples were subjected to Fourier transform infrared (FTIR) spectrometer complemented by 525 x-ray diffraction (XRD) to assess the mineralogical composition of the soil samples from the study 526 527 area. The FTIR absorption bands for the different soil samples with their corresponding minerals are 528 presented in Table 5. The FTIR results showed that among the four minerals identified, quartz and kaolinite were the major minerals based on their peak intensities in all the samples (Fig. 7). Quartz is 529 abundant mineral in soil due its chemical structure and hardness, it gives characteristics Si-O 530 symmetrical stretching vibration at 778 - 796 cm⁻¹, Si-O asymmetrical bending vibrational frequency 531 at 462 - 694 cm⁻¹ and Si-O asymmetrical stretching vibration at 1082 - 1162 cm⁻¹ because of low Al 532 to Si substitution (Paramasivam et al., 2019). The chemical weathering of Feldspars produced 533 Kaolinite (Al₂Si₂O₅(OH)₄) which is a clay mineral. The presence Kaolinite is indicated with a 534 vibrational frequency in the range 3621.75 - 3695 cm⁻¹ and 911.39 - 1004.29 cm⁻¹. The appearance of 535 peaks in the range 2859.8 - 2916.9 cm⁻¹ and 2915.9 - 2921.6 cm⁻¹ indicated the presence of organic 536 carbon and a sharp peak between 3612 - 3896 cm⁻¹ revealed the presence of carbonate (Fig. 6). 537

538 **3.7.2 XRD analysis**

539 XRD analysis was applied to complement the FTIR results. The diffraction patterns are obtained 540 through phase identification by comparing the diffraction pattern with a JCPDS database. The 541 minerals identified were quartz and kaolinite (Fig. 7). Quartz showed a high peak intensity in all the 542 samples analyzed. As a result of disordered (loss of crystalline nature) of the respective minerals, 543 some minerals did not show in the XRD results as compared to the results obtained from the FTIR 544 (Fig. 6). Nevertheless, the XRD results also shows that quartz and kaolinite were the major minerals 545 in the soil samples with high peak intensities (Fig. 7) and this is in tandem to FTIR results.

546

3.9 Pollution source identification and apportionment using multivariate statistical analysis 548 Multivariate statistical analysis such as Pearson's correlation analysis (CA), principal component 549 analysis (PCA) and hierarchical cluster analysis (HCA) were employed to evaluate the 550 interrelationship among the PTMs, radioisotopes and its associated radiological parameters in 551 dumpsites soil characteristics due to its usefulness as a dimension reduction technique widely applied 552 553 in environmental monitoring studies without losing much information (Gbadamosi et al., 2018a). The 554 statistical analyses were carried out using the commercial statistics package IBM SPSS statistics (version 26). 555

3.9.1 Pollution sources identification and apportionment using principal component analysis (PCA)

Principal component analysis (PCA) was carried out to apportion the potentially toxic metals 558 (PTMs), radioisotopes and their associated radiological parameters pollution sources in these study 559 areas. This was performed based on varimax orthogonal rotation and eigenvalue > 1 after Kaiser-560 meyer Olkin Normalization measuring of adequacy and Bartlett's test significant value for all the 561 variables. Potentially toxic metals and radioisotopes and their associated radiological parameters 562 originating from similar source(s) were always grouped with strong loadings. The results obtained 563 showed that there is an initial dimensional reduction of all the dataset to four components which 564 explained 86.8 % of the data variation (Table 6). The first principal component (PC-1) explained 61.7 565 % of the total variance and was heavily loaded by ²³⁸U and all the radiological parameters and health 566 hazard parameters (0.814-0.998) (Table 6, Fig. 8). This confirmed that the radiological and health 567 hazards parameters were mainly determined by the level of ²³⁸U in the samples. The second principal 568 component (PC-2) was dominated by the toxic metals Ni, Cd, Pb and Cr with high loadings values of 569 0.768, 0.724, 0.632 and 0.577 respectively (Table 6). This component which accounted for 10.0% of 570 the total variance determined and is mainly attributed to anthropogenic sources of the toxic metals in 571 572 the dumpsites. This component could be defined as a combination of natural and anthropogenic

component. The third and fourth principal components (PC-3 and PC-4) accounted for 8.8 % and 6.3 573 % of the total variance explained. PC-3 loaded heavily on 40 K (0.874) while PC-4 loaded heavily by 574 Zn and moderately negative with ²³²Th respectively (Table 6). From the results and values obtained 575 from pollution assessment, it clearly revealed that PC-1 and PC-2 represent mixed natural, 576 anthropogenic and lithogenic sources mainly from aggregation of lead-containing materials in the 577 waste on the dumpsites, vehicular emission, industrial discharges and atmospheric depositions (Kong 578 et al., 2014; Li et al., 2018), while PC-3 and PC-4 are mainly related to anthropogenic activities 579 resulting from agronomic used of potassium-rich fertilizer and waste discharge from nearby 580 industries into the dumpsites. 581

582 Table 6: Principal component analysis (after varimax rotation) showing contribution of 583 statistically dominant variables measures in this study

	Pri	Communalities			
Variables	1	2	3	4	_
Hext	0.998	0.054	0.022	0.030	0.999
Raeq (BqKg ⁻¹)	0.998	0.054	0.022	0.030	0.999
AEDEin (mSvyr ⁻¹)	0.991	0.051	0.112	0.045	0.999
D _R (nGyhr ⁻¹)	0.991	0.051	0.112	0.045	0.999
ELCRout	0.991	0.051	0.112	0.045	0.999
AEDEout (mSvyr ⁻¹)	0.991	0.051	0.112	0.045	0.999
ER (μ Rhr ⁻¹)	0.990	0.051	0.118	0.032	0.998
Iγr	0.986	0.050	0.147	0.032	0.997
AGDE (mSvyr ⁻¹)	0.982	0.049	0.169	0.045	0.998
Hint	0.963	0.060	-0.182	0.086	0.971
AUI	0.886	0.062	-0.439	0.008	0.981
²³⁸ U (BqKg ⁻¹)	0.814	0.061	-0.416	0.145	0.860
Ni (mg/kg)	0.092	0.768	0.000	-0.031	0.599
Cd (mg/kg)	0.069	0.724	-0.363	0.186	0.695
Pb (mg/kg)	0.282	0.632	0.280	0.311	0.654
Cr (mg/kg)	-0.300	0.577	0.120	-0.418	0.611

⁴⁰ K (BqKg ⁻¹)	0.411	-0.004	0.874	0.069	0.939
Zn (mg/kg)	0.100	0.115	0.030	0.818	0.693
²³² Th (BqKg ⁻¹)	0.380	0.023	-0.400	-0.409	0.472
Eigen-value	11.833	1.863	1.699	1.068	-
% of variance explained	61.7	10.0	8.8	6.3	-
Cumulative (%)	61.7	71.7	80.5	86.8	-

Component Plot in Rotated Space



585



587 **3.9.2** Hierarchical cluster analysis (HCA)

Hierarchical cluster analysis (HCA) is a multivariate statistical analysis commonly used to quantify the similarity of the variables and analyze the effect of the factor on the samples using a variance approach to define the distance between the clusters (Jiang et al., 2015). In HCA, the average linkage method along with coefficient distance was used with an explicit dendrogram (Fig S3). In the dendrogram shown, the potentially toxic metals (PTMs), radioisotopes and their associated radiological parameters were grouped into four statistically significant clusters. Cluster-I consisted of (all the heavy metals, H_{ext}-Iγr-H_{int}-AUI-²³²Th-D_R-AEDE_{out}-²³⁸U-Ra_{eq}). These are considered to
originated from mixed natural, anthropogenic and lithogenic sources. However, the anthropogenic
influences are of greater impact. Cluster-II is made up of (ER-ELCR_{out} and AEDE_{in}). Cluster-III
consisted of AGDE while Cluster-IV consisted of ⁴⁰K. Cluster-II and cluster-III have a minor
similarity and a major similarity with Cluster-IV (Fig. S3). This shows that Cluster-IV (⁴⁰K) was the
main determinant of the radiological parameters such as ER, ELCRout, AEDE and AGDE showed in
Cluster-III and Cluster-III.

601 **3.9.3** Pearson's correlation coefficients analysis

Pearson's correlation matrix (PCM) was carried out in order to establish the strength of association
and direction of the linear relationship between the variables through the calculation of the linear
Pearson product moment correlation coefficient (r).

A correlation analysis between radioisotopes and their associated radiological parameters, potential 605 toxic metals in the sampling areas clearly revealed a strong positive correlation between ²³⁸U and the 606 estimated associated radiological parameters at 0.01 level of significance ($P \le 0.01$) (2-tailed) (Table 607 S5). A moderate correlation occurs between ⁴⁰K and the estimated radiological parameters. This 608 shows that ²³⁸U activity concentration is the main factor responsible for the radiological parameters 609 determined in the samples. A weak correlation existed between ²³²Th with other radioisotopes. A 610 moderate positive correlation existed between Pb and Ni (0.374), Cd and Ni (0.470) at 0.01 level of 611 significance ($p \le 0.01$) (Table S5). All other PTMs correlated weakly with other radioisotopes and 612 their associated radiological parameters in this study. This correlation results vividly revealed that the 613 calculated radiological parameters correlated highly in the study, due to high concentration of ²³⁸U 614 and ⁴⁰K in the dumpsites soil samples, which can be attributed to their common anthropogenic 615 sources of pollution through extensive fertilizer application and radioactive materials in the deposited 616 617 waste.

618 3.10 Spatial distribution of radioisotopes

The results obtained for the activity ratio for ²³⁸U:²³²Th activity concentrations in the dumpsite soils 619 samples showed that ²³⁸Th activity concentrations were about 4 times higher than the ²³²Th activity 620 concentrations in all the soil samples. The activity concentration of ⁴⁰K is about 42 times and 11 621 times higher than the ²³²Th and ²³⁸U in the soil samples. The ratio ²³⁸U/²³²Th ratio (Table S6) is 622 higher than the world's average value of unity in 92 % of the samples (fifty-five sampling points). 623 The ratio of ²³⁸U: ²³²Th, ²³⁸U: ⁴⁰K and ²³²Th: ⁴⁰K suggests that the soil samples from a certain area 624 had higher or lower ²³⁸U, ²³²Th and ⁴⁰K activity concentration to be economically suitable for 625 extraction. ⁴⁰K and ²³⁸U activity concentrations were higher in all the samples which may be due to 626 (i) the presence and aggregation of clinical or medical wastes containing radioactive materials 627 (uranium) in the dumpsites and extensive use of potassium rich fertilizer like phosphate in the soils. 628 The sum and range of the individual activity ratio of the radioisotopes used for the box-plot in Fig. 629 S4, shows that for ${}^{238}U/{}^{232}$ Th, ${}^{238}U/{}^{40}$ K and 232 Th/ 40 K the range of the activity ratios are 1.65 - 244.6, 630 0.03 - 25 and 0.002 - 5.19 respectively (Fig. S4). This shows that the activity ratio for $^{238}U/^{232}Th$ 631 were higher than the stipulated range of 0.7 - 0.4 (Mitchell et al., 2002; ICRP, 1976) and for some 632 sampling sites, ²³²Th/⁴⁰K also recorded higher values (Table S6). This could indicate the possibility 633 of ²³⁸U extraction from the study areas. 634

635 4 Conclusion

The activity concentrations of the radioisotopes and their associated radiological parameters, pollution and health risk assessment of the PTMs from the selected dumpsites in Ijebu-Ode were evaluated. The mean activity concentrations of the radioisotopes in the dumpsite soil samples were of the order; 40 K > 238 U > 232 Th. The total and average activity concentrations of the radioisotopes 238 U and 40 K were higher than the reported global average values. The dumpsites in these locations are hazardous based on the values of the average and maximum radiological and health parameters such

D_R, Raeq, Hext, Hint, Ivr, ER, AEDEindoor, AEDEoutdoor and ELCRoudoor which were higher than the world 642 average values in most of the sampling points. The mean and maximum concentrations of the PTMs 643 were below the maximum acceptable limit (MAL) for the countries considered except Cd which has a 644 higher than the MAL in all the countries. The pollution and ecological risk assessment showed that 645 the selected dumpsites are moderately-strongly polluted with the metals and that the dumpsites could 646 647 pose great ecological risks to the biomes. The results of the health risk assessment model revealed that the carcinogenic risk for children and adults with CR values of the three carcinogenic metals (Cr, 648 Cd and Ni) higher than the acceptable range $(1 \times 10^{-6} \leftrightarrow 1 \times 10^{-4})$. Mineralogical composition of the soils 649 was obtained from FTIR and confirmed through XRD suggest that the major minerals are quartz and 650 kaolinite with high peak intensities. Multivariate statistical analysis applied to attributes the sources 651 of radioisotopes and potentially toxic metals (PTMs) established an interdependent relationship 652 between the variables. This study therefore comprehensively evaluated and apportioned the PTMs 653 and radioisotopes pollution sources, human exposure risk assessment to the toxic metals from the 654 selected dumpsites and will serve as a reliable baseline data for future monitoring of the study areas 655 and areas of similar geology and environmental usage. It also provides useful information for the 656 Federal and State Ministry of Environment on waste management with respect to the continuous 657 deleterious emissions from waste dumpsites to the surrounding environment and likely contamination 658 659 of proximate surface and ground waters.

- 660 **Conflict of interest**
- 661 The authors declare no conflict of interest.

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663 Acknowledgements

Authors wish to express appreciations Tai Solarin University of Education (TETFUND-IBR-Team 6)grant and Babcock University for providing the financial support through Babcock University

666 Research Grant No: BU/RIIC/2018/016 to support this project. The authors also acknowledge the 667 technical support received from Mrs Owoade and Mr Olubunmi Emmanuel Tope of the National 668 Institute of Radiation and Protection, University of Ibadan, Ibadan during the analysis. All 669 anonymous reviewers of this manuscript are well appreciated by the authors.

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