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ORIGINAL ARTICLE

Radiometric evaluation of excessive lifetime cancer probability due to naturally occurring radionuclides in wastes dumpsites soils in Agbara, Southwest, Nigeria

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Abstract The concentration and spatial distribution of the gamma ray emitting ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides in dumpsite soils in Agbara were analyzed with the aim of evaluating the radiation hazards and excessive lifetime cancer risk using well calibrated HPGe γ-ray spectrometry technique. The ranges of activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs are 11.5 ± 1.0 ↔ 166 ± 40 Bq kg^{-1}, 15.6 ± 1.8 ↔ 31.4 ± 2.3 Bq kg^{-1}, 20.4 ± 1.3 ↔ 366 ± 30 Bq kg^{-1} and 0.52 ± 0.1 ↔ 8.44 ± 0.2 Bq kg^{-1} respectively. Radiological parameters such as absorbed dose rate, radium equivalent, annual effective dose equivalent, internal and external hazard indices, gamma level index, activity utilization index, annual genetic significant dose equivalent, exposure rate and excessive lifetime cancer risk were calculated to know the complete radiological hazardous nature of the dumpsite soils to the inhabitants of the sites. The calculated radiological parameters were higher than the world average value in two of the sampling points. The ratio of the detected radioisotopes was calculated for spatial distribution of natural radionuclides in the study area. RESRAD computer code was applied to calculate the total effective dose equivalent (TEDE). The code was also used to calculate the probability of excess lifetime cancer incurred by dwellers/inhabitants of the dumpsites, the level of which was determined to be 0.5×10^{-4} and 2.5×10^{-5} for Idowale and Ibijola dumpsite soils over a period of 30 years respectively. Therefore, the radiological risks to the general populations from waste enhanced naturally occurring radioactive materials (WENORM) from the Idowale dumpsite top soils are considered to be significant.

1. Introduction

There has long been concern about the issue of soil pollution by radionuclides and heavy metals because of their severity of toxicity for plant, animal, human beings, environment and

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their lack of biodegradability in the environment. Soil is a primary recipient of many of the waste products and chemical used in modern society. Soil is the primary reservoir of radionuclides and other pollutants in the atmosphere, hydrosphere and biota, and thus plays a fundamental role in the overall nuclide cycle in nature (Cao et al., 2010). Soil is an important environmental media that sustain life, one of the ways soil can be polluted is through improper disposal of hazardous wastes. Radioactive materials in soil pose potential threats to the environment and can damage human health through various absorption pathways such as direct ingestion, dermal contact, and diet through the soil–food chain, inhalation, and oral intake (Lu et al., 2011).

Human activities create wastes and the way these are handled, stored, collected and disposed can give rise to impacts on the environment and public health (Ademola et al., 2015). Hazardous waste can cause pollution, damage to health and even death. The environmental problem posed by solid wastes which are improperly disposed in most cities and town has been of concern to federal, state local and community development authorities in Nigeria. Poor waste management poses several challenges to the well-being of city residents due to the potential of the waste to pollute water, food sources, land, air and vegetation (Njoroge, 2007), when the wastes are not properly managed or disposed (Porteous, 1985).

Hazards posed by such waste dumpsite are not only in term of odor and presence of disease causing micro-organism, but can arise from the radiation emanating from such dumpsite (Ojoawo et al., 2011), which occur as results of accumulation and reaction of different radioactive materials in the waste indiscriminately dumped on open waste site. At waste dumpsites, there are possibilities for radiation to be emitted due to the presence of radioactive waste in the landfills as well as naturally occurring radionuclides in the soil. The radioactive contamination of soil, water and air can be transferred to humans through the soil via plants (^{40}K) or through inhalation (^{222}Rn and ^{220}Rn). These radionuclides even at low concentrations can have potential impacts on the environmental quality and human health and may pose a long term risk (Ademola et al., 2015). Moreover, risk assessment which is an effective scientific tool will enable decision makers to manage sites so contaminated in a cost-effective manner while preserving public and ecosystem health (Zhao and Kaluarachchi, 2002). Therefore, objectives of this study were to determine the concentration of waste enhanced naturally occurring radionuclides (WENORM) present in representative soil samples from some selected waste dumpsites in Agbara by gamma-ray spectrometry in order to estimate the, excessive lifetime cancer probability, radioactivity disequilibrium and other radiological hazard indices from these dumpsites to the general public and provide a reliable baseline data for future radionuclide evaluation in the area.

2. Material and methods

2.1. Description of the study area

Agbara which is an industrially populated town in Ogun State is located within latitude 6°20′0″ N and 6°35′0″ North of the Equator and longitude 3°5′0″ E and 3°10′0″ East of the Greenwich Meridian (Fig. 1). The area stands on a low-lying gent undulating terrain with altitude ranging between 30 and 80 m above sea level. The area is characterized by high annual temperature, high rainfall, high evapotranspiration and high relative humidity which make it to be classified as humid tropical region (Akanni, 1992). The prevailing wind direction was Southwesterly at the period of study (wet season period–Jun e–October) while the prevailing wind speed ranged range between 2.52 to 3.55 m s^{-1}. The prevailing wind direction in the dry season (November–March) is Northeasterly. Two major municipal dumpsites soils of about 3 m thickness and covered an area of 10,000 m^2 were used and they are Ibijola dumpsite along Ibijola hospital way and Idowale dumpsite (major) along Idowale road dumpsite.

2.2. Sample coding

The soil samples were coded as follows in order to prevent identification error. The sample codes consist of four alphabets. The first alphabet stands for the study area (Agbara), the second alphabet stands for representative soil samples (A, & B), and the last two alphabets stands for the soil layer type e.g. TS which connotes Top Soil and SS which connotes Sub Soil. All soil samples were controlled with a control sample different from the above stated soil samples, this was done in order to know the contamination level and dose rate of radionuclide in these dumpsite soils compared to that of the dumpsite free soil (control soil samples) that was collected at about 40 km from Idowale dumpsite within the same geological formation with Ibijola dumpsite. AA stands for Idowale dumpsite soils while AB stands for Ibijola dumpsite soils.

2.3. Sample collection and preparations for radiochemical analysis

A total of ten (10) soil samples were obtained from the two major dumpsites in the study area and a waste free control samples within the same geological formation with the two selected dumpsites using a hand-driven stainless steel soil auger. Firstly with a garden rake, the waste was removed to expose the soil under the waste dumps from where samples were collected. Five sets of soil samples were collected from each of the dump sites (Idowale and Ibijola). At each location, the dumpsite was divided into two sections. In each section, five soil samples were collected and composited to obtain a representative samples using coning and quartering technique. This implies that five composite or representative samples were obtained from each dumpsite under investigation. The soil samples were taken at about 0–15 cm depth (using a meter rule) by the use of stainless steel hand-driven soil auger, and taken to the laboratory in labeled polythene bags stored and were air-dried at laboratory temperature in order to avoid cross contamination or pollution of the samples. The samples were then pulverized by grinding, and filtered through a 2 mm mesh sieve. Two hundred grams (200 g) of pulverized soil samples was subsequently measured using an analytical weighing balance with a precision of ±0.01 g and packed into cylindrical containers (beaker) which were carefully labeled. These samples were safely conveyed to Natural Institute of Radiation Protection and Research, University of Ibadan, Ibadan, South-West Nigeria, at the laboratory the plastic were hermitically sealed with adhesive tape and kept for minimum of

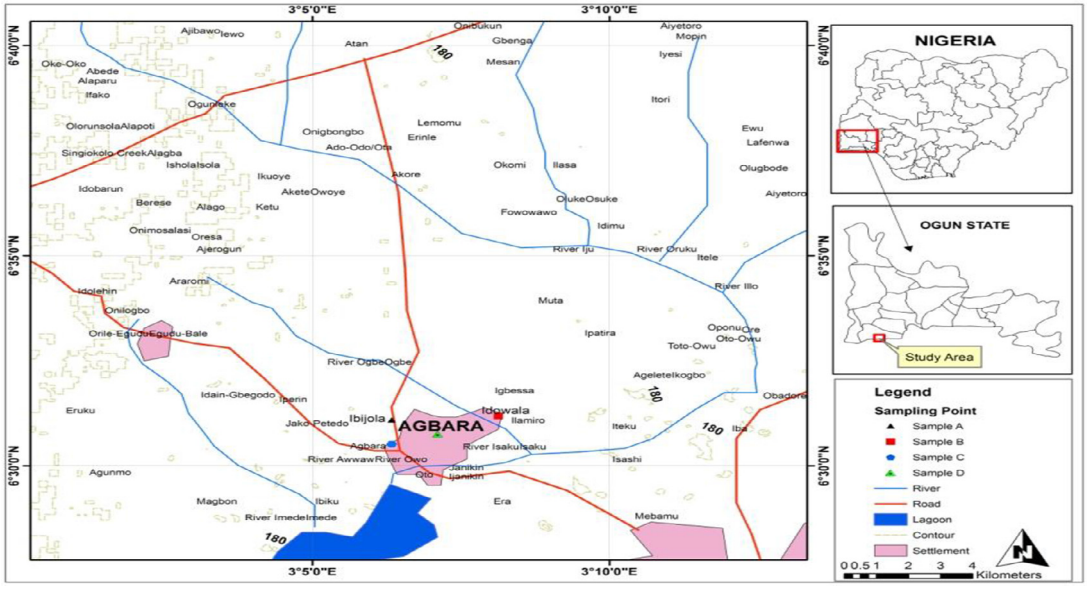

Fig. 1 Map of the study area showing sample locations.

30 days to ensure that the parent and daughter nuclide in the sample were at secular equilibrium between radium and its gaseous decay progenies. At the end of the four weeks in-growth period, the samples were subjected to gamma-ray spectroscopy counting.

3. Radioactivity measurements

3.1. System used for measurements

Activity concentration ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs were measured by high resolution, low background γ-ray spectrometer equipped with a vertical coaxial high purity germanium (HPGe) detector with a relative efficiency of 20.2%. The spectrometer was tested for its linearity and calibrated for energy and efficiency using the well calibrated standard gamma source obtained from an International Atomic Energy Agency (IAEA), laboratories, Vienna, Austria (Ademola et al., 2008). Efficiency is the measure of percentage of radiation at a given detector detect from the overall yield that is emitted from the source into a solid angle of usually 4π in the photopeaks (Hossain et al., 2012). Accuracy of efficiency calibration of detector is necessary to obtain the high precision measurements with radioactive samples. The resolution of the detector is 7.5% at 0.662 MeV of ^{137}Cs. This resolution is capable of distinguishing the gamma ray energies of interest in the study. All the samples were counted for 36,000 s in order to obtain good statistics for ^{238}U, ^{232}Th, ^{137}Cs and their daughter products and ^{40}K. Also measurements were repeated at intervals for quality assurance purposes as well as to ascertain the stability of the measuring system. The background radiation due to the naturally occurring radionuclides in the environment around the detector was measured using an empty plastic container; the empty plastic container was measured in the same manner as the soil samples for the same counting time of 36,000 s (10 h). The background spectrum was subtracted from the measure spectra to obtain the net radionuclides activities. The background, reference sample and the soil samples were measured under the same conditions (Jibiri et al., 2014). The γ-ray photo-peaks corresponding to ^{214}Pb at (242.0 keV, 295.2 keV & 351.9 keV), ^{214}Bi (609.3 keV, 768.4 keV, 806.19 keV, 1120.3 keV, 1377.669 keV and 1401.516 keV respectively) and ^{234}Pa (1001.025 keV) were considered for identifying ^{238}U. The γ-ray photo-peaks of ^{228}Ac at (209.3 keV, 338.3 keV, 409.5 keV and 911.1 keV), ^{208}Ti at (277.4 keV, 583.2 keV and 860.6 keV), ^{212}Bi at (727.33 keV and 785.37 keV), ^{212}Pb (238.6 keV and 300.1 keV and ^{224}Ra (240.986 keV) were used to identify ^{232}Th in the samples. The radioisotopes ^{40}K and ^{137}Cs were estimated from emits gamma ray with energy of 1460.8 keV and 661.65 keV. Hence the determination of ^{40}K and ^{137}Cs were considered as direct (Sam and Abbas, 2001). The activity concentration (A_c) of ^{238}U, ^{232}Th and ^{40}K in Bq kg^{-1} was obtained using the relation in Eq. (1) (Amrani and Tahtat, 2001):

$$\text{Sample activity}(A_C)(\text{Bq kg}^{-1}) = \frac{C_i}{\varepsilon(E)P_\gamma(E)tm} \quad (1)$$

where C_i is the net peak area after subtraction of background of the gamma-ray line at energy E, ε (E) is the detector efficiency of such gamma-ray line at photopeak energy (E), $P_\gamma(E)$ is the emission probability of the gamma-ray photons of energy (E) under consideration, t is the time of measurement in seconds and m is the mass of the sample in kg. The below detectable limit (BDL) of each radionuclide was determined from the background radiation spectrum for the same counting time for the dumpsites soil samples.

The detection limits (DL) required to estimate the minimum detectable activity in a sample were obtained using Eq. (2)

$$LLD(\text{Bq kg}^{-1}) = 4.65\frac{\sqrt{C_b}}{t_b}f \quad (2)$$

where C_b is the net background count in the corresponding peak t_b is the background counting time (s), and f is the factor

that converts cps (counts per second) to activity concentration (Bq kg^{-1}).

3.2. Radiation hazard parameters

In order to estimate the radiation hazards incurred by the population due to the activity levels of the measured waste enhanced naturally occurring radionuclides (WENORM) in the selected dumpsite soils in Agbara Industrial town, some radiation hazard indices were calculated and the details of calculations are given in Table 2. These are used to reassess the statistical information about excessive lifetime cancer risk (ELCR), γ-ray absorbed dose rate (D_R), outdoor and indoor annual effective dose equivalent ($AEDE_{outdoor}$ & $AEDE_{indoor}$), Annual gonadal dose equivalent (AGDE), radium equivalent (Ra_{eq}), activity utilization index (AUI), exposure rate (ER), internal and external hazard indices (H_{int} & H_{ext}) and external (γ-radioactivity) level index ($I_{\gamma r}$) for the present study. Even though the total activity concentration of radionuclides is calculated, it does not provide the exact indication about the total radiation hazards due to uneven distribution of the waste enhanced naturally occurring radionuclide (^{40}K, ^{238}U and ^{232}Th) in the soil. Statistics of all the calculated radiological parameters and their recommended levels by UNSCEAR (2000) are given in table 3. Distributions of the above radiological parameters are shown in Figs. 2 and 3.

4. Results and discussion

The results obtained from NIRPR shows different activity concentration of the radionuclides as shown below.

4.1. Activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs

Table 2 shows the activity concentration of naturally and artificially occurring radioactive elements ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs determined in the dumpsite soil samples. The activity concentration of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in the dumpsite soils ranged from $11.5 \pm 1.0 \leftrightarrow 166 \pm 40$ Bq kg^{-1}, $15.6 \pm 1.8 \leftrightarrow 31.4 \pm 2.3$ Bq kg^{-1}, $20.4 \pm 1.3 \leftrightarrow 366 \pm 30$ Bq kg^{-1}, and $0.52 \pm 0.1 \leftrightarrow 8.44 \pm 0.2$ Bq kg^{-1} respectively. In comparison with the world average value, mean activity concentration of radionuclides ^{238}U is higher than the world average value of 33 (UNSCEAR) and it is about 5 to 6 times higher than this recommended limit in two sampling locations (AATSI and AATS2) (Fig. 2), whereas ^{232}Th, ^{40}K and ^{137}Cs average values were below the recommended average value (UNSCEAR, 2000). It was observed from the results that the activity concentration values are in the order $^{137}Cs < {}^{232}Th < {}^{238}U < {}^{40}K$ in all sampling sites (Fig. 2). Similar observation has been reported by (Ademola et al., 2015). Higher activity concentrations of the radionuclides were obtained in the top soil samples from Idowale dumpsites (Fig. 2). The control sample recorded the lowest activity concentration of the radionuclides analyzed. Higher concentration of ^{40}K might be due to the higher silica content which is one of the abundant mineral deposits in the soil (Navarrete et al., 2014).

4.2. Radiological parameters

Radium equivalent (Ra_{eq}) which is used to describe the γ-output from different mixtures of ^{238}U, ^{232}Th and ^{40}K in the soil samples from the study area and it is calculated using equation in Table 1. The calculated values of Ra_{eq} ranged from $36.6 \pm 3.7 \leftrightarrow 235 \pm 46$ Bq kg^{-1} with a mean value of 85.5 ± 10 Bq kg^{-1}. The values from all the sampling sites are below the maximum permissible level of 370 Bq kg^{-1}(UNSCEAR, 2000). The absorbed dose rate (D_R) which is the energy imparted per unit weight of the irradiate material is also calculated using the equation in Table 1 and results shown in Table 2. The absorbed dose rate (D_R) values varied from $16.2 \pm 1.6 \leftrightarrow 109 \pm 21$ nGy hr^{-1} with a mean value of 38.7 ± 5.0 nGy hr^{-1} (Table 2). From the present study, it is clearly revealed that the mean absorbed dose rate (D_R) was below the recommended limits of 55 nGy hr^{-1} (UNSCEAR, 2000) but higher by a factor of 2.0–2.4 times in two of the sampling points (AATS1 and AATS2) (Fig. 4).

Table 1 Summary of the radiological parameters of all the dumpsite soil samples.

S/N	Radiological parameters	Units	Used formula[a,b]
1	Absorbed dose rate (D_R)	nGy hr^{-1}	$D_R = (0.462A_U + 0.604A_{Th} + 0.0417A_K)$
2	Radium equivalent (Ra_{eq})	Bq kg^{-1}	$Ra_{eq} = (A_u + 1.43A_{Th} + 0.077A_k)$
3	External Hazard index (H_{ext})	–	$H_{ext} = A_U/370 + A_{Th}/259 + A_K/4810 \leqslant 1$
4	Internal Hazard index (H_{int})	–	$H_{int} = A_U/185 + A_{Th}/259 + A_K/4810 \leqslant 1$
5	Annual effective dose equivalent ($AEDE_{outdoor}$)	μSv yr^{-1}	AEDE (outdoor) = $D_R \times 8766$ h $\times$ 0.7 Sv/Gy $\times$ 0.2 $\times 10^{-3}$
6	Annual effective dose equivalent ($AEDE_{indoor}$)	μSv yr^{-1}	AEDE (indoor) = $D_R \times 8766$ h $\times$ 0.7 Sv/Gy $\times$ 0.8 $\times 10^{-3}$
7	Annual gonadal dose equivalent (AGDE)	μSv yr^{-1}	$AGDE = 3.09A_U + 4.18A_{Th} + 0.314A_K$
8	Gamma level index ($I_{\gamma r}$)	–	$I_{\gamma r} = A_U/300 + A_{Th}/200 + A_K 3000 \leqslant 1$
9	Activity utilization index (AUI)	–	$AUI = A_U/50 + A_{Th}/50\ f_{Th} + A_K/500 f_K \leqslant 2$
10	Exposure rate (ER)	μR hr^{-1}	ER (μR hr^{-1}) = $1.90\ A_U + 2.82\ A_{Th} + 0.179\ A_K$
11	Excess lifetime cancer risk (ELCRoutdoor)	–	ELCR = AEDEoutdoor $\times$ DL $\times$ RF

[a]UNSCEAR (2000) and [b]Al-Trabulsy et al. (2011), where A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in (Bq kg^{-1}) present in waste dumpsites soil respectively. f_U (0.462), f_{Th} (0.604) and f_K (0.0417) are the fractional contributions to the total dose rate due to γ-radiation from the actual radionuclide of ^{238}U, ^{232}Th and ^{40}K, respectively. DL and RF is duration of life (70 years) and risk factor (S v^{-1}), fatal cancer risk per sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public.

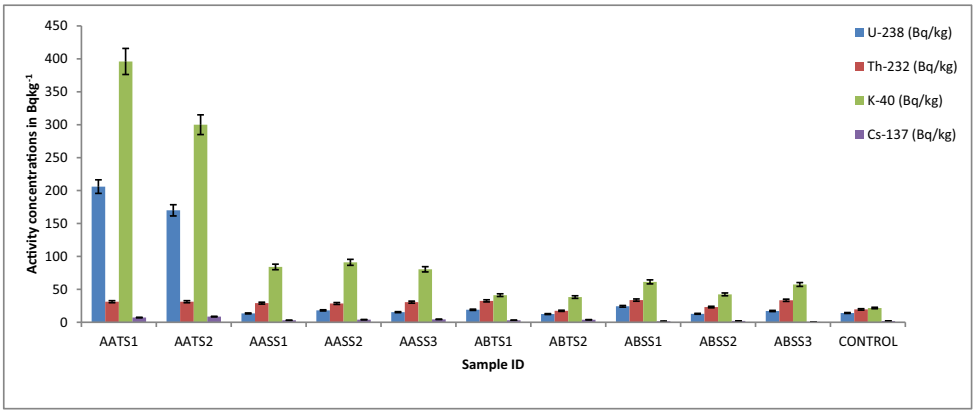

Fig. 2 Activity concentration of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs.

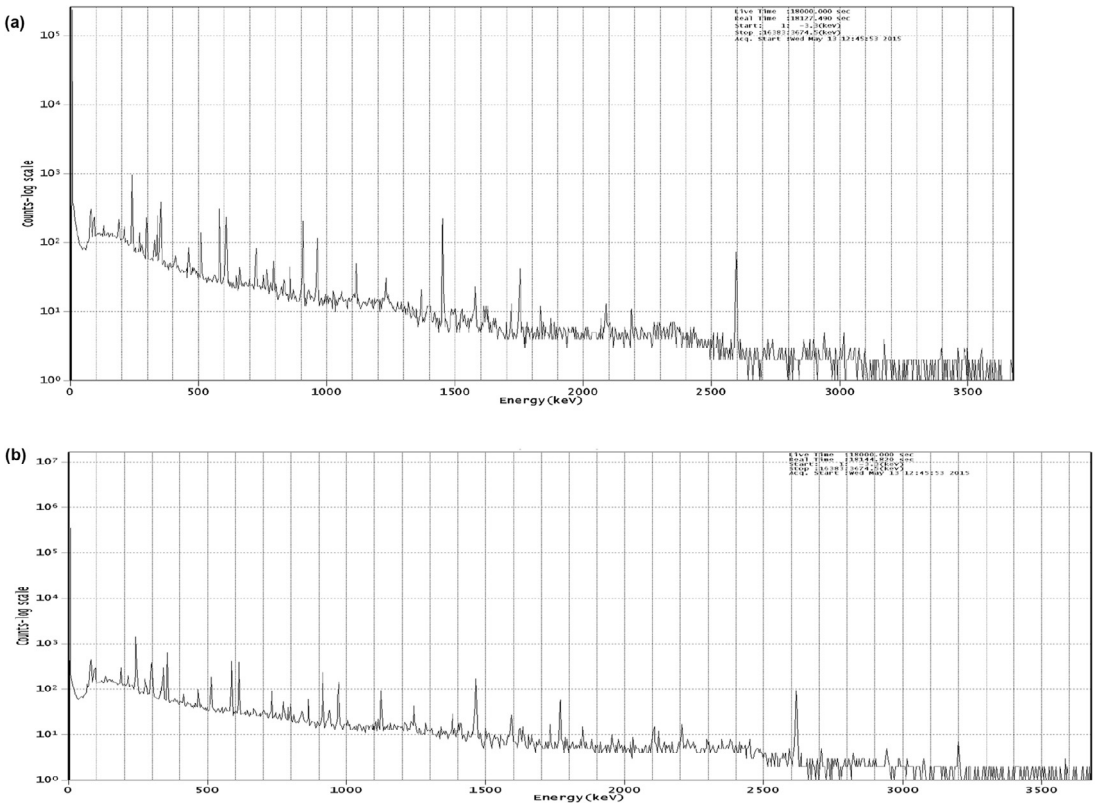

Fig. 3 (a) Showing the gamma-ray spectrum of Sample AASS1 (Idowale soil sample). (b) Showing the gamma-ray spectrum of Sample ABSS1 (Ibijola soil sample).

The calculated AEDE values ranged from 19.9 ± 1.7 ↔ 134 ± 26 μSv yr^{-1} with a mean value of 47.4 ± 6.1 μSv yr^{-1}. This is below the world average value of 70 μSv yr^{-1} (UNSCEAR, 2000). Higher and the second higher values of AEDE were obtained at AATS1 and AATS2 (134 ± 26 and 115 ± 20 μSv yr^{-1}) (Fig. 4) which are about 2 times higher than the recommended level. Only these two locations have the AEDE values above the world average value, this is due to a high activity concentration radionuclide distribution in the top-soil samples. The calculated values for the rate of exposure (ER) of individuals and scavengers to these radionuclides in the selected dumpsites was below the maximum limit of (600 μR hr^{-1}) in all samples. Only two sample points i.e. AATS1 and AATS2 have an exposure rate value of 462 ± 88 μR hr^{-1} and 398 ± 67 μR hr^{-1} which are moderately high but below the recommended limits of 600 μR hr^{-1} in all samples. This shows that the exposure limit to this ionizing radiation by the people around the dumpsites are insignificant in 80% of samples.

The annual genetically significant dose equivalent (AGSDE) or annual gonadal dose equivalent (AGDE) is a measure of the genetic significance of the yearly dose equivalent received by the population's reproductive organs (gonads) (Morsy et al., 2012; Ramasamy et al., 2014). Not all living cells

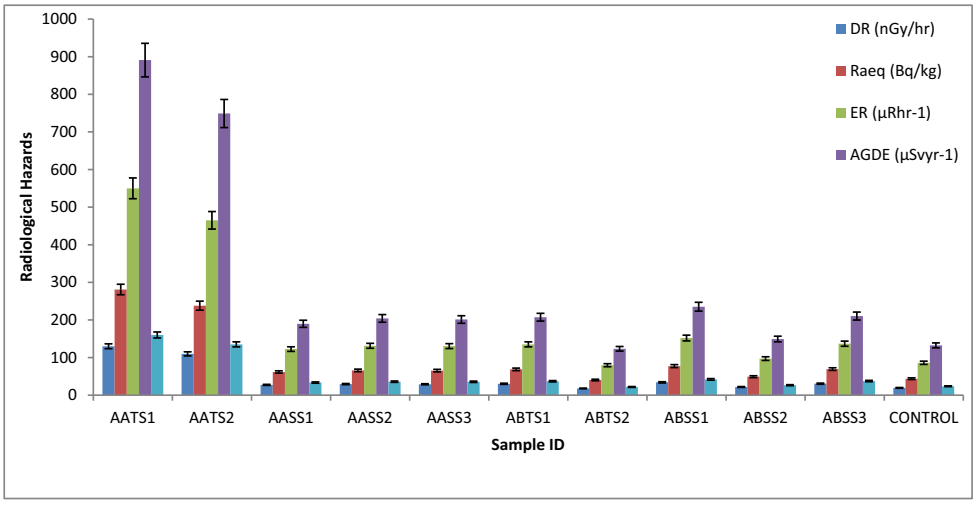

Fig. 4 Radiological parameters.

are equally sensitive to radiation. Those cells which are actively reproducing are more sensitive than those which are less or not reproducing (Ramasamy et al., 2013). A direct interaction of radiation with reproductive cells could result in the death or genetic mutation of the cell, whereas a direct interaction with the DNA of a dormant cell could have less effect (Ramasamy et al., 2013). The gonads, bone marrow and bone surface are considered to be the organs of importance since they are active (UNSCEAR, 2000). Hence the AGSDE or AGDE due to the activities of ^{238}U, ^{232}Th and ^{40}K was calculated and is ranged from 112 ± 9.4 ↔ 748 ± 143 μSv yr^{-1}, with an average value of 265 ± 33 μSv yr^{-1} (Table 3). This present average value is less than the recommended level (1000 μSv yr^{-1}) (UNSCEAR, 2000). However, two sampling points (AATS1 and AATS2) (Fig. 4) have higher contribution from ^{40}K and ^{238}U with moderately higher AGDE values among the sampling locations.

4.3. Internal, external hazard & activity utilization index (H_{int}, H_{ext} & AUI)

In order to assess the health effects from human exposure to the ionizing radiation of the earth's surface materials containing ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radiation hazards from the internal and external sources are important. The prime objective of these hazard indices is to limit the radiation dose equivalent limit of 1 μSv yr^{-1} (Ramasamy et al., 2013). Detailed statistics for H_{int} & H_{ext} calculations are given in Table 2. The index value must be less than unity in order to keep the radiation hazard to be insignificant. The calculated external hazard values are between 0.10 ± 0.01 ↔ 0.63 ± 0.12 with an average value of 0.23 ± 0.03 (Table 3). The average value of external hazard index (H_{ext}) is less than unity. Hence, there is no radiological risk emanating from external exposure of the inhabitant or people working/living around the dumpsites to these deleterious natural radionuclides. The calculated values of internal hazard index (H_{int}) ranged from 0.13 ± 0.01 ↔ 1.09 ± 0.23(Fig. 4) with an average value of 0.34 ± 0.05. The value of internal hazard exceeds the recommended limit in two of the sampling points i.e. AATS1 and AATS2. Therefore the soils in these two areas pose an internal radiological risk to the inhabitants and people working or living around the study locations owing to the harmful effects of the ionizing radiation from the harmful radioactive substances in the wastes soils.

The calculated values for activity utilization index (AUI), which can be used to determine the possibility of using the wastelands soil for building construction ranged from 0.30 ± 0.03 (ABTS2) ↔ 1.91 ± 0.04 (AATS1) with an average of 0.70 ± 0.09 (Table 4). This calculated average value is lower than the world average of 2 (UNSCEAR, 2000). Exceedance of the recommended upper limit is noted in two sampling points (AATS1 & AATS2), with the highest value of 1.91 ± 0.04 observed at (AATS1). These higher AUI values in these two locations can be attributed to the higher concentration of radionuclides in these sites. Here also, average relative contribution of the γ-index is due to higher ^{40}K followed by the contributions due to ^{238}U and ^{232}Th.

4.4. Excessive lifetime cancer risk (ELCR)

The potential carcinogenic and mutagenic effects are characterized by evaluating the probability of cancer incidence in a population of individuals for a specific lifetime from projected intakes cum exposure with chemical-specific dose response data (i.e. slope factors). Excessive lifetime cancer risk is the probability or risk for an individual to develop a cancerous cell due to an exposure to toxic and deleterious substances from a multiple exposure pathways over time. Excessive lifetime cancer risk (ELCR) was calculated using the equation presented in Table 1 and the calculated values ranged from 0.07 ± 0.006 × 10^{-3} ↔ 0.47 ± 0.09 × 10^{-3} with an average value of 0.17 ± 0.02 × 10^{-3} (Table 3). The present average ELCR value is less than the world average value of (0.29 × 10^{-3}) (UNSCEAR, 2000) (Table 3) (Fig. 5). Two (2) of the sampling points (AATS1 and AATS2) (20%) have the ELCR values higher than the world average value. These results show that the lifetime cancer risk due to exposure through the use of this dumpsite soils for a maximum duration of 70 years is high in these two locations. Hence, the subsequent use of this dumpsite soil for building, construction, agriculture or for soil remediation studies and other purpose should be discouraged.

Table 2 Geological location and activity concentration of radionuclides (A_c) & absorbed dose rate value for different soil samples.

Sample ID	Latitude	Longitude	Activity Concentrations (Bq kg^{-1})				D_R (nGy hr^{-1})	Ra_{eq} (Bq kg^{-1})
			^{238}U	^{232}Th	^{40}K	^{137}Cs		
AATS1	06°11.140′N	03°52.735′E	166 ± 40	28.8 ± 2.5	366 ± 30	7.13 ± 0.1	109 ± 21	235 ± 46
AATS2	06°11.151′N	03°33.618′E	140 ± 30	29.2 ± 2.0	277 ± 23	8.44 ± 0.2	93.9 ± 16	203 ± 35
AASS1	06°11.140′N	03°52.735′E	12.9 ± 0.6	27.1 ± 2.0	79.5 ± 4.6	2.83 ± 0.2	25.6 ± 1.7	57.8 ± 3.8
AASS2	06°11.213′N	03°41.636′E	17.0 ± 1.1	26.3 ± 2.2	86.0 ± 5.0	3.93 ± 0.1	27.3 ± 2.1	61.2 ± 4.7
AASS3	06°11.186′N	03°48.596′E	14.6 ± 0.8	28.1 ± 2.5	76.1 ± 4.4	4.39 ± 0.1	26.9 ± 2.1	60.6 ± 4.7
ABTS1	06°11.562′N	03°31.614′E	17.8 ± 1.2	29.9 ± 2.6	38.8 ± 2.4	2.94 ± 0.2	28.0 ± 2.2	63.6 ± 5.1
ABTS2	06°11.729′N	03°21.641′E	11.5 ± 1.0	15.6 ± 1.8	36.1 ± 2.2	3.82 ± 0.1	16.2 ± 1.7	36.6 ± 3.7
ABSS1	06°11.638′N	03°10.558′E	22.2 ± 2.1	31.4 ± 2.3	58.0 ± 3.4	1.31 ± 0.3	31.6 ± 2.5	71.6 ± 5.7
ABSS2	06°11.729′N	03°21.641′E	12.0 ± 0.9	21.0 ± 2.1	40.0 ± 2.4	1.89 ± 0.2	19.9 ± 1.8	45.1 ± 4.1
ABSS3	06°11.562′N	03°15.526′E	16.3 ± 0.9	31.1 ± 2.3	54.1 ± 3.3	0.52 ± 0.1	28.6 ± 1.9	64.9 ± 4.4
Control	–	–	13.5 ± 0.6	17.9 ± 1.7	20.4 ± 1.3	1.93 ± 0.1	17.9 ± 1.34	40.7 ± 3.1
Min.	–	–	11.5 ± 1.0	15.6 ± 1.8	20.4 ± 1.3	0.52 ± 0.1	16.2 ± 1.6	36.6 ± 3.7
Max.	–	–	166 ± 40	31.4 ± 2.3	366 ± 30	8.44 ± 0.2	109 ± 21	235 ± 46
Range	–	–	155 ± 39	15.8 ± 0.5	346 ± 29	7.92 ± 0.1	92.8 ± 19	198 ± 42
Mean			40.3 ± 7.2	26.0 ± 2.2	103 ± 7.5	3.56 ± 0.2	38.7 ± 5.0	85.5 ± 10
World Average			33[a]	45[a]	420[a]		55[a]	370[a]

[a]UNSCEAR (2000); [b]Al-Trabulsy et al. (2011)).

More so, people working or living around the dumpsite should avoid the use of these soils for any of the aforementioned purposes.

4.5. Spatial distributions

The investigation of ^{238}U:^{232}Th activity concentrations in the dumpsite soils revealed that ^{238}U activity concentrations are seen to be of 6 times higher than the ^{232}Th activity concentration in the measured soil samples from AATS1 (Idowale dumpsite top soils). The ratio $^{238}U/^{232}Th$ ratios (Table 5) were higher than the world's average value of 1 in the two top soil samples from Idowale waste dump sites. The activity concentration of ^{40}K shows that on average ^{40}K are about 2 and 12 times higher than ^{238}U and ^{232}Th activity concentration in the collected soil samples. This ratio ^{232}Th: ^{238}U, ^{238}U: ^{232}Th, ^{232}Th: ^{40}K and ^{238}U: ^{40}K gives an indication that the samples from a certain region or part have higher uranium and potassium than thorium concentrations to be economical feasible for the ^{238}U and ^{40}K extraction. On the average, activity concentration of ^{40}K and ^{238}U is high in all the samples which may be due to (i) the presence of loamy and clay soils couple with extensive use of potassium rich fertilizer like phosphate in the soils. The $^{232}Th/^{238}U$ ratio was also calculated (Table 5), from these results, the ratio were higher than the stipulated range 0.7–0.4 mentioned by (Mitchell et al., 2002; ICRP, 1976) for igneous zircon sources in all the soil samples. All the soil samples of the study area have $^{232}Th/^{238}U$ ratio above this range. This could be indicated that the present study confirmed the highest presence of xenolith zircon in the soil samples.

4.6. Determination of annual doses & probability of excess cancer risk lifetime using Residual radiation software (RESRAD)

Residual Radioactivity software (RESRAD, version 6.5, 2013) an onsite computer code developed by the environmental assessment of the Argonne National laboratory was used to assess and predict the total effective dose equivalent (TEDE) and probability of excess lifetime cancer risk incurred by workers (Fouzey et al., 2013), scavengers and people living around the dumpsites who are exposed to the deleterious waste enhanced naturally occurring radioactive materials (WENORM) ^{238}U, ^{232}Th ^{40}K and ^{137}Cs emanated from the selected dumpsites. The derivation for the dumpsite clean-up or remediation criteria depends on the sum of the fraction rule from multiple contamination from ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides (Eq. (3)), This sum of rule fraction from the multiple contamination must be less than unity and if the calculated value from (Eq. (3)) is greater than 1, there is need for soil remediation in order to reduce the cancer risk. The deepest depth of the dumpsites contamination in this present study was determined to be 2 m.

From this scenario, the potential exposure route examined include (i) direct exposure to external γ-radiation from the contaminated materials in the dumpsites (ii) internal radiation from inhalation of radionuclides (Radon) polluted dust by the workers, scavengers and people leaving around the dumpsites and (iii) the internal γ-radiation from ingestion of contaminated soil. While water and food materials is assumed to be from other sources and not been produced from the dumpsites. The simulation was performed using RESRAD software for a period 70 years and that waste management officer, scavengers and people around the selected dumpsites have 3 h daily exposure to these substances from the dumpsites. Finding from the RESRAD results revealed that the probability that the workers and people living around Idowale and Ibijola dumpsites might be affected by cancer through their multiple route of exposure to this ionizing radiation is 0.5×10^{-4} and 2.5×10^{-5} as shown in (Figs. 6 and 7 respectively).

Sum of the fraction rule equation (SOFRE) (Fouzey et al., 2013)

$$\mathrm{SOFRE} = \left[\frac{IR_U}{SR_{(U)tmax}} + \frac{IR_{Th}}{SR_{(Th)tmax}} + \frac{IR_K}{SR_{(K)tmax}} + \frac{IR_{Cs}}{SR_{(Cs)tmax}}\right] \leqslant 1 \quad (3)$$

Table 3 Radiological parameters of Dumpsites Soils in Agbara.

Sample ID	ER ($\mu R\ hr^{-1}$)	AGDE ($\mu Sv\ yr^{-1}$)	H_{ext}	H_{int}	$I_{\gamma r}$	AUI	AEDEout ($\mu Sv\ yr^{-1}$)	ELCR $\times 10^{-3}$ outdoor
AATS1	462 ± 88	748 ± 143	0.64 ± 0.12	1.08 ± 0.2	0.82 ± 0.2	1.91 ± 0.4	134 ± 26	0.47 ± 0.09
AATS2	398 ± 67	641 ± 108	0.55 ± 0.09	0.93 ± 0.2	0.71 ± 0.1	1.67 ± 0.3	115 ± 20	0.40 ± 0.07
AASS1	115 ± 7.6	178 ± 11.7	0.16 ± 0.01	0.19 ± 0.01	0.21 ± 0.02	0.45 ± 0.03	31.5 ± 2.1	0.11 ± 0.01
AASS2	122 ± 9.2	190 ± 14.2	0.17 ± 0.01	0.21 ± 0.02	0.22 ± 0.02	0.48 ± 0.04	33.5 ± 2.5	0.12 ± 0.01
AASS3	121 ± 9.4	187 ± 14.3	0.16 ± 0.01	0.20 ± 0.02	0.22 ± 0.02	0.48 ± 0.04	33.0 ± 2.5	0.12 ± 0.01
ABTS1	125 ± 10	192 ± 15.3	0.17 ± 0.01	0.22 ± 0.01	0.22 ± 0.02	0.53 ± 0.04	34.2 ± 2.7	0.12 ± 0.01
ABTS2	72.3 ± 7.4	112 ± 11.3	0.10 ± 0.01	0.13 ± 0.01	0.13 ± 0.01	0.30 ± 0.03	19.9 ± 2.0	0.070 ± 0.01
ABSS1	141 ± 11	218 ± 17.2	0.19 ± 0.02	0.25 ± 0.02	0.25 ± 0.02	0.59 ± 0.05	38.8 ± 3.1	0.14 ± 0.01
ABSS2	89.2 ± 8.1	137 ± 12.3	0.12 ± 0.01	0.15 ± 0.01	0.16 ± 0.01	0.37 ± 0.03	24.2 ± 2.2	0.085 ± 0.008
ABSS3	128 ± 8.8	197 ± 13.4	0.02 ± 0.01	0.22 ± 0.01	0.23 ± 0.02	0.53 ± 0.04	35.1 ± 2.4	0.12 ± 0.008
Control	79.6 ± 6.2	123 ± 9.37	0.11 ± 0.001	0.15 ± 0.01	0.14 ± 0.01	0.34 ± 0.03	22.0 ± 1.7	0.077 ± 0.006
Min.	72.3 ± 7.4	112 ± 9.4	0.10 ± 0.01	0.13 ± 0.01	0.13 ± 0.01	0.30 ± 0.03	19.9 ± 1.7	0.07 ± 0.006
Max.	462 ± 88	748 ± 143	0.63 ± 0.12	1.09 ± 0.23	0.82 ± 0.16	1.91 ± 0.40	134 ± 26	0.47 ± 0.09
Range	390 ± 81	636 ± 134	0.53 ± 0.11	0.96 ± 0.22	0.69 ± 0.15	1.61 ± 0.37	114 ± 24	0.40 ± 0.08
Mean	168 ± 21	265 ± 33	0.23 ± 0.03	0.34 ± 0.05	0.30 ± 0.04	0.70 ± 0.09	47.4 ± 6.1	0.17 ± 0.02
World average 600[a]	1000[a]	≤1[a]	≤1[a]	≤	0.5[a]	≤2[a]	70[a]	0.29[a]

[a]UNSCEAR (2000); [b]Al-Trabulsy et al. (2011).

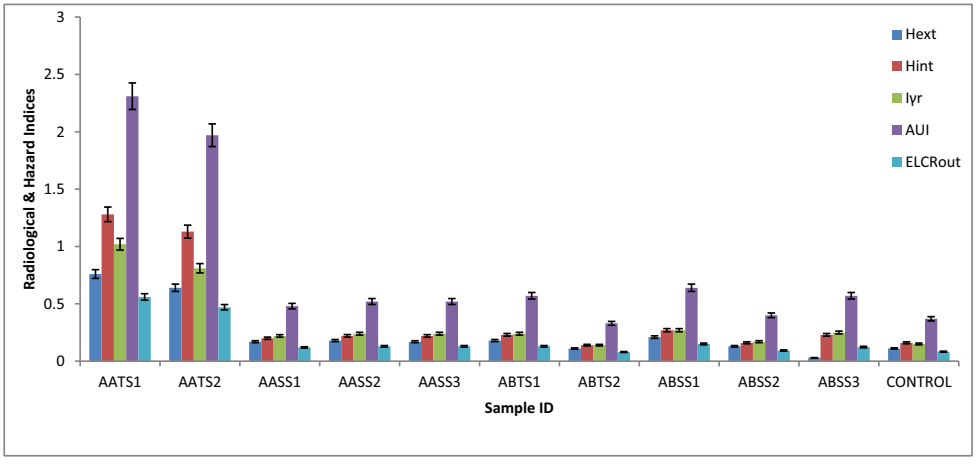

Fig. 5 Radiological parameters & hazard indices.

Table 4 Comparison of activity concentration and radiological hazard indices with those found in similar studies.

Sample	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	D_R (nGy hr^{-1})	Ra_{eq} (Bq kg^{-1})	References
Control	13.45 ± 0.57	17.87 ± 1.66	20.39 ± 1.32	17.86 ± 1.32	40.57 ± 3.05	Present study
Agbara	42.95 ± 7.87	26.84 ± 2.20	111.05 ± 7.98	40.69 ± 5.31	89.49 ± 11.67	Present study
Lagos	69.19 ± 19.10	14.49 ± 3.22	409.44 ± 86.08	57.80 ± 14.36	121.44 ± 30.33	Oladapo et al. (2012)
Sango Ota	122.10 ± 20.60	3.0 ± 1.23	3.30 ± 9.8	58.36 ± 10.67	126.64 ± 23.11	Ademola et al. (2014)
Ojota (Lagos)	23.1 ± 2.5	35.1 ± 2.1	318.9 ± 27.4	48.8	97.8	Ademola et al. (2015)
Ado Ekiti	36.57 ± 2.70	25.73 ± 5.60	758.51 ± 132.93	64.07 ± 10.17	131.77 ± 20.94	Isinkaye and Faweya (2006)
Ibadan (Oritaperin)	27.93 ± 10.52	44.93 ± 7.24	488.91 ± 217.24	60.43 ± 7.41	92.26 ± 5.60	Jibiri et al. (2014)
Osogbo	52 ± 6	22 ± 2	186 ± 6	45.07 ± 4.23	97.78 ± 9.32	Faweya and Babalola (2010)
Port Harcourt	41.96 ± 5.53	62.61 ± 18.97	643.10 ± 5.94	84.02 ± 14.26	181.01 ± 33.12	Avwiri and Olatubosun (2014)
Crustal average	35	35	370	59	370	UNSCEAR (2000)

The ^{238}U, ^{232}Th and ^{40}K activity concentrations, Ra_{eq} and D_R for the samples from the terrestrial environment were compared with the values found in a similar study (Table 4).

where IR_U, IR_{Th} and IR_K are Initial radionuclide concentrations ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in the soil in (pCi/g) and $SR_{(U)t}$, $SR_{(Th)t}$ and $SR_{(K)t}$ are the concentration of the single radioactivity ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs (pCi/g) after a particular time t-maximum (70 years in this scenario). This Sum of fraction rule equation value must be <1 for the soil not to

Table 5 Activity ratios.

S/N	Sample ID	$^{238}U/^{232}Th$	$^{238}U/^{40}K$
1	AATS1	6.58	0.52
2	AATS2	5.45	0.57
3	ABTS1	0.58	0.46
4	ABTS2	0.72	0.33
5	AASS1	0.46	0.16
6	AASS2	0.64	0.20
7	AASS3	0.50	0.19
8	ABSS1	0.72	0.39
9	ABSS2	0.56	0.31
10	ABSS3	0.52	0.30
11	Control	0.72	0.65

be considered for remediation processes (Fouzey et al., 2013). The calculated SOFRE value for Idowale and Ibijola dumpsite soils is 0.90 and 0.66 respectively. Idowale dumpsite soils have higher SOFRE value than Ibijola dumpsite soils. Indication of this is that Idowale dumpsite soils have greater tendency of causing harms to the inhabitants and it will be due for remediation process in the nearest future since the SOFRE value is almost a unity (0.9). Though both locations SOFRE value falls within the stipulated value by nuclear regulatory commission (NRC). The total effective dose equivalent (TEDE) received by the people or animal living or working around the dumpsites was calculated using Eq. (4).

$$\text{TEDE} = \text{SOFRE} * 0.25\ \text{mSv yr}^{-1} \tag{4}$$

where 0.25 mSv yr^{-1} is the basic dose limit adopt by nuclear regulatory commission (NRC, 2015) criteria and 0.3 mSv yr^{-1} by (IAEA, 2011). The calculated TEDE values for Idowale and Ibijola dumpsites are 0.225 and 0.166 mSv yr^{-1}. This revealed that the TEDE value from the selected dumpsites soils in Agbara is below the regulatory dose limit of 0.25 mSv yr^{-1} set by (NRC, 2015) and 0.3 mSv yr^{-1} set by (IAEA, 2011). Though, the Idowale dumpsite soils have higher SOFRE and TEDE values that are almost equal to the allowable limit. The implications of this, are that the dumpsites soils from Idowale dumpsite cannot be used for any purposes and a complete recommendation of the soil is thereby recommended.

5. Conclusion

The mean activity concentrations of the naturally and artificial occurring radionuclides ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in all the soil samples are of this order $^{40}K > {}^{238}U > {}^{232}Th > {}^{137}Cs$. The total and average activity concentrations of the radionuclides (^{238}U and ^{40}K) were higher than the reported worldwide average values. Radiological parameters like absorbed dose rate, $I_{\gamma r}$, H_{int}, $AEDE_{outdoor}$ and $ELCR_{outdoor}$ were higher than the recommended level in two of the samples from Idowale dumpsites, while other radiological parameters (AGDE, AUI, H_{ext}, ER, and Ra_{eq}) fall below the worldwide average values.

The soils of Idowale and Ibijola dumpsites pose little radiological threat to the people working/living around the dumpsites in which the cumulative effects can be detrimental to human health if the exposure is for a longer period of time. This finding was also corroborated with the Residual Radioactivity (RESRAD computer code) which revealed a moderately high cancer probability and SOFRE value for Idowale dumpsite soils than Ibijola dumpsite soils The total effective dose equivalent (TEDE) value was lower than the regulatory dose limit of 0.25 mSv yr^{-1} and 0.3 mSv yr^{-1} set by (NRC, 2015) and (IAEA, 2011) respectively. Though, Idowale dumpsite has a moderately high value. Thus the dumpsite soils from Idowale dumpsite pose a significant threat to the waste management staff and other inhabitants of this study area and remediation procedure is therefore recommended for future use of the soils samples from this study locations.

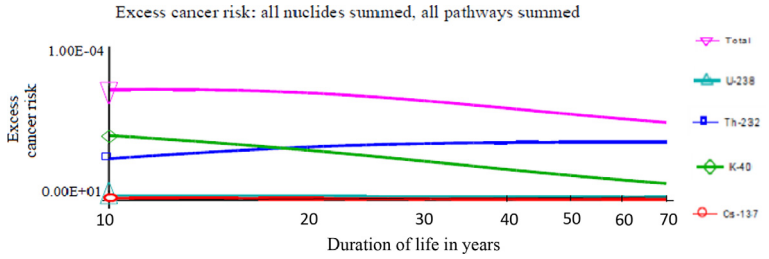

Fig. 6 Showing the probability of excessive cancer lifetime incurred by the people living around Idowale dumpsite.

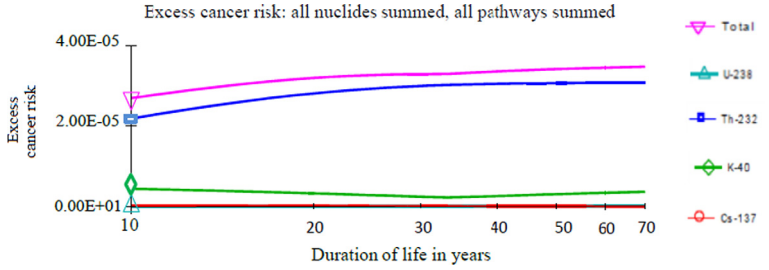

Fig. 7 Showing the probability of excessive cancer lifetime incurred by the people living around Ibijola dumpsite.

Conflict of interest

The authors have no conflict of interest.

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