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Evaluation of radon exhalation rate and excessive lifetime cancer risk in Dumpsites in Ondo city Southwestern Nigeria

E.B. Faweya[1,*], G.O. Olowomofe[1], H.T. Akande[1], O. Faweya[2], G.E. Adesakin[1]

[1]*Department of Physics, Faculty of Science, Ekiti State University, Nigeria*
[2]*Department of Statistics, Faculty of Science, Ekiti State University, Nigeria*

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Corresponding authors:
E.B. Faweya, PhD.,

E-mail:
febdeprof@yahoo.co.uk

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ABSTRACT

Background*:** Ondo city in Ondo State Nigeria includes Ondo East and West could be described as a research city due to antecedent number of research institutes. ***Materials and Methods*:** Dose due to radioactivity content of soil samples from sixteen waste dump sites in Ondo, Southwestern Nigeria was determined using gamma-ray spectrometer in order to evaluate the radiation hazard health profile and excess lifetime cancer risk ELCR. ***Results: The weighted average value of radon emanation coefficient, mass exhalation rate, concentration in soil gas and surface exhalation rate were 10.5%, 15.2 µBq kg^{-1} s^{-1}, 2.2 kBq m^{-3} and 18.8 mBq $m^{-1}s^{-1}$. The weighted mean concentrations obtained were 354 ± 53, 58±14 and 23 ± 2Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. Radium equivalent activity (119.11Bq kg^{-1}), Outdoor external dose (54.26 nGy h^{-1}), Indoor external dose (107.04 nGy h^{-1}), and total average annual effective dose (0.59 mSv y^{-1}) were obtained. From the measured γ-rays spectra, elemental concentrations were determined for ^{232}Th (mean 5.8ppm), ^{226}Ra (mean 4.8ppm) and ^{40}K (mean 1.1 %). Thorium was lower than world average 7.4ppm by a factor of 1.3; radium was higher than the world average (2.8 ppm) by factor 1.7 while potassium was relatively equal to world average (1.3 %). ***Conclusion:*** The total excess lifetime cancer risk ELCR was found to be 1.95 x 10^{-3} which was slightly higher than the world average. In addition, a good correlation was found between the radium concentration and radon exhalation rate in the area.

Keywords: *Activity concentration, dose, dumpsites, excess lifetime cancer risk ELCR, radon parameters.*

INTRODUCTION

The great interest expressed worldwide for study of naturally occurring radiation and environmental radioactivity has led to the interest of extensive survey in many countries. This is done in order to gain information about the present levels of harmful pollutants such as wastes discharged to the environment itself or the living creatures [1, 2]. Out of the three naturally radon isotopes, ^{222}Rn which is chemically inert is the most used as tracer for studying global pollution and climate change owing to its suitable half-life [3, 4] and therefore need to be estimated in the study area. Radon atoms located within solid grains are unlikely to become available for release and contaminate the atmosphere, owing to their very low diffusion coefficients in solids. However, if they are located in the interstitial space between grains such as soil samples, they may diffuse to the surface. Radon release to the earth's surface

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may take place in form of transportation-diffusion and adventive flow cause the movement of the emanated radon atoms through the residue or soil profile to the ground surface, emanation-radon atoms formed from the decay of radium escape from the grains (mainly because of recoil) into the interstitial space between the grains and exhalation-radon atoms that have been transported to the ground surface and then exhaled to the atmosphere [5, 6]. Therefore, information on distribution of ^{222}Rn from earth's surface is considered as useful for identifying areas with a risk of radon exposure to public. Transfer of ^{222}Rn from soil to the atmosphere involves some of the same processes controlling the soil to air exchange of important greenhouse gases such as CH_4, CO_2 and NO_2. Since ^{222}Rn is chemically inert, it does not undergo complicated chemical reactions and its source is ^{226}Ra in the soil [7].

The process of discharge and burning of untreated or incompletely treated wastes in the city may generate gaseous ^{222}Rn and particulate emission such as fly ash that contains ^{226}Ra, ^{232}Th and ^{40}K will hasten the deterioration of receiving air, soil and water bodies. The high water table, and sandy soil of Ondo could allow toxins to leach easily into the ground water which is a major source of drinking water and concentration of radionuclides may reach hazardous levels. External exposure is caused by gamma-rays emitted by aforementioned radionuclides, while internal exposure is caused by α and β particles due to inhalation of the radioactive inert gas radon and its short-lived progenies [8, 9]. Data on the amounts of dose received by populace from soil in municipal solid waste MSW discarded in the city are still not known. Consequently, the risks associated with the disposal of municipal solid waste MSW to landfill in the city have not been fully elucidated. Dumpsites are a major source of radioactivity that contributes significantly to the background level of radiation [10]. Knowledge of radon emanated in dumpsites is thus important because data emanating from cancer registries in Nigeria is scanty and not being population based. This study aims to (i) determine the concentrations of naturally occurring radionuclides ^{226}Ra, ^{232}Th, ^{40}K and ^{222}Rn in the dumpsites of Ondo city metropolis in Ondo State, (ii) evaluate the radiation health hazard indices, (iii) determine excess lifetime cancer risk ELCR for the local population and visitors from dose from the action of aforementioned radionuclides.

MATERIALS AND METHODS

Study Area

Ondo metropolis is located in an area of mean elevation of about 300m above sea level and lies between latitudes 7°05′20″ and 7.088923^0 north of the equator and longitudes 4°47′57″ and 4.7990935° east of the Greenwich meridian. The climatic conditions are 26°C, wind W at 5 km hr^{-1} and 82 % relative humidity. Ondo city fell within Benin-Owena River basin; a region that experiences mean annual evapotranspiration 4622.8 mm in any normal year and an estimated mean annual precipitation 6604 mm [11]. Use of radionuclide in medicine, industry and research are on the increase in Nigeria. There are many centres using radiation facilities in the city such as Wesley University of Science and Technology, Adeyemi College of Education, Ondo City Polytechnic, Ondo State university of Medical Sciences, Techville, The National Institute for Education Planning and Administration (NIEPA) in collaboration with UNESCO, Gani Fawehinmi Diagnostics centre and other hospitals. To date, it is the economic nerve centre of Ondo state and the second largest and most populated city in the state with population of 358,430 [12]. It has the highest concentration of manufacturing industries in the state and the trade centre for the surrounding region. Plants such as yams, cassava, cocoa, grain and tobacco are cultivated. Cotton is also cultivated and is used to weave cloth called Aso Oke fabric. Wastes from these industries, house wastes such as electronic waste (e-waste), couple with attendance number of the aforementioned research institutes and hospitals account for the magnitude of waste generated in the metropolis.

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Sampling

Soil samples were collected from sixteen dump sites located within Ondo city during the periods of January-March. The city was divided into four (4) zones such as, zone 1 Ondo-Ore road 18 samples, zone 2 Ondo- Akure road 14 samples, zone 3 Ondo-Okelaje road 17 samples and zone 4 Ondo-Ife road 16 samples as seen in figure 1. Four sites that are five (5) to ten (10) years above in each zone were studied. Studied sites were chosen because lag period of 10 years is generally assumed for cancers to develop as a result of cancer-inducing agents such as ionizing radiation and 5 years for lymphatic and hematopoietic cancers [13]. From each site, 3-5 soil samples were collected from an area of 0.5 m x 0.5 m up to depth of 25 cm. In this way, control samples were collected from undisturbed and non-eroded sites without any influence of man- made structures to ensure that samples were representative of the sites from where they were taken. They were dried crushed to small grain size passed through a sieve of 1.0 mm grid size and 250 g of each sample was packed into a hermitically sealed plastic container of 60 mm height by 65 mm diameter. The samples were sealed for a minimum of 30 days before counting to re-establish the radioactive equilibrium between ^{226}Ra and its daughter's products due to the possible escape of radon gas during handling.

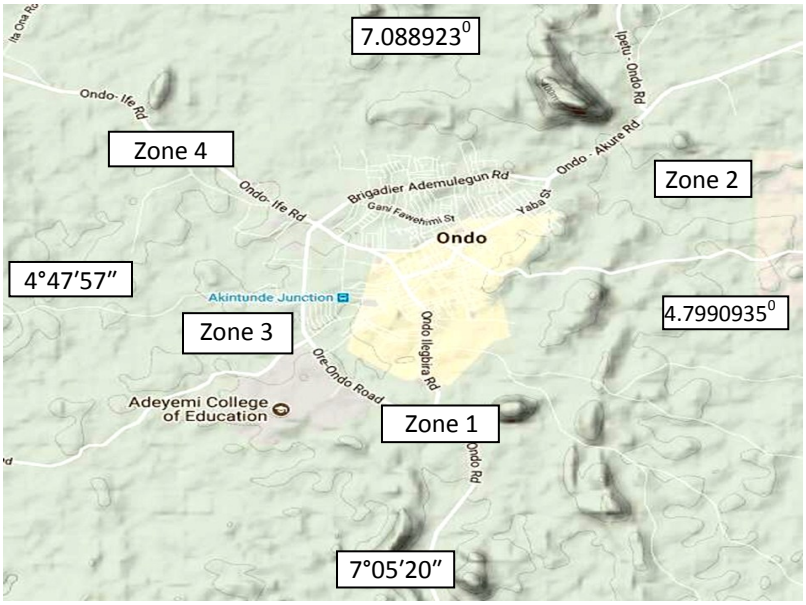

Figure 1. Modified Ondo Google Satellite Map Showing Zones of Sample Collection. Map data ©2017 Google (14)

Radioactivity Measurements

Counting and Measurements

Gamma counting was done using a lead-shielded 76mm × 76mm NaI (TI) detector crystal (by Canberra Inc. USA) coupled to Canberra series 10 plus Multichannel Analyzer (MCA) through a preamplifier. The detector had a resolution of about 8% at energy of 0.662 MeV. This was enough to distinguish the gamma ray energies being of interest in the present study. The choice of radionuclides to be detected as a reference was made based on the fact that the NaI (TI) detector used in this study had a poor energy resolution. Hence the photons emitted by them would be sufficiently discriminated if their emission probability and their energy were high enough, and the surrounding background continuum low enough. Therefore, the activity concentration of ^{214}Bi determined from its 1.765 MeV γ–ray peak was chosen to provide an estimate of ^{226}Ra in the samples, while that of the daughter radionuclide ^{208}TI determined from its 2.615 MeV γ–ray peak was chosen as an indicator of ^{232}Th, while ^{40}K was determined by

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measuring the 1.460 MeV γ–rays emitted during the decay of ^{40}K. Finally, ^{137}Cs was also quantified using 0.662 MeV γ–ray peak. An empty container was counted for the same period as the counting time for samples (10 hrs) to access the background concentrations of the γ -rays. The activity concentration level in each of the samples was calculated after measurement and subtraction of the background counting using the equation (1) [15].

$$C(Bq\ kg^{-1}) = \frac{CPSX100X100}{I\ X\ \epsilon_{ff}\ X\ m} + \frac{CPS_{error}X100X100}{I\ X\ \epsilon_{ff}\ X\ m} \quad (1)$$

Where *CPS* is Net count rate per second, *I* is Branching intensity, ϵ_{ff} is Efficiency of the detector, *m* is sample mass in kg and CPS_{error} is Standard Deviation of net count rate per second. Sample used is traceable to source reference 375 from IAEA laboratories (Siebersdorf, and Vienna, Austria). The lowest limits of detection (LLD) of the activities of the natural radionuclides in each sample were determined using environmental measurement laboratory procedure [16] using the equation (2).

$$DL\ (Bq\ kg^{-1}) = \frac{1.96}{Kem}\left(\frac{B}{T} + SD_b{}^2\right)^{\frac{1}{2}} \quad (2)$$

Where *B* is the background count, SD_b is the estimated standard error of the net background count rate in the peak, *T* is the counting time (s), ε is the counting efficiency (CPs/Bq), *m* is the mass of the sample, *k* is the factor that converts Cps (count per second) to Bq and 1.96 represents the 95% confidence level. The LLD values obtained were 17.03 Bq kg^{-1}, 5.09 Bq kg^{-1} and 5.06 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. The activity values below LLD were considered to be below detection limit (BDL) of the detector.

Soil Porosity

Sample collection for porosity

Cylindrical steel ring of dimension 5cm × 5 cm manufactured in the Mechanical Workshop of Department of Physics, Ekiti State University was used for the collection of the sample. It was driven to the ground by a block of wood and hammer in 3-5 sampling points in the sixteen sites. This was done to reduce error in measurements. The ring was removed with care by excavating the surrounding soil with shovel and all the roots and pebbles from the bottom were cut-off. The core soil in the ring was carefully transferred into plastic bag. The samples were then transferred to laboratory for analysis. At the laboratory, the samples were air-dried at room temperature. Thereafter, samples were dried within 15 minutes on high for 3 times until constant weight was obtained to ensure that all the pores in the samples have been drained of water. The microwave oven used was Prestige (from Meyer Intellectual Limited, UK) model CS25-U, input rating 1400W, output rating 800W, voltage 230-240 AC, frequency 2450 MHz, Kesa UK, HU1 3AU. Dried samples were transferred to premeasured beakers to measure the volume. Water from measuring container close to the samples was added until the samples became saturated. Volume of water used to saturate each sample was recorded by subtracting the volume of water that was left from the initial volume of water. This is equal to the pore volume in each sample. The porosity was determined using the following equation (3)

$$porosity(P_t) = \frac{pore\ volume\ (V_p)}{solid\ volume(V_s) + pore\ volume(V_p)} \quad (3)$$

The average porosity was calculated as 46.7 %. The average Bulk density was determined using the porosity for each sample and true particle density 2.65 kg m^{-3}. The average Bulk density was 1.4 kg m^{-3}.

Radon parameters

Emanation coefficient which is also referred to as emanating power is defined as the fraction of radon atoms generated that escape the solid phase in which they are formed and become free to migrate through the bulk medium. Radon concentration and exhalation rate were measured using gamma spectrometry technique which is a useful non- destructive method [5]. Another set of thirty two samples (two from each site) sealed in hermitically plastic containers were measured three (3) times using gamma rays emitted from ^{214}Bi before and after establishment of radioactive equilibrium

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between radon and its progenies. The emanation coefficient is calculated using the following equation (4) [5].

$$\mathcal{E} = \frac{CPS_{eq} - CPS_0}{CPS_{eq}} \quad (4)$$

Where Ɛ is the emanation coefficient, CPS_{eq} is the specific counts selected from the peak of ^{214}Bi in equilibrium condition and CPS_o is the counts corresponding to in initial condition. The value of Ɛ determined for the soil samples ranged from 3.6 to 20 % with an average of 10.5 %.

Radon mass exhalation rate was estimated using the following equation (5) as defined by [1].

$$R_M\,(Bq\,kg^{-1}s^{-1}) = \lambda_{Rn} * C_{Ra} * \mathcal{E} \quad (5)$$

Where λ_{Rn} is the decay constant of radon given as (2.1×10^{-6} s^{-1}), C_{Rn} is the activity concentration of ^{226}Ra in the soil samples and Ɛ is the ^{222}Rn emanation coefficient of the soil samples.

The radon concentration in soil gas C_{Rn} without radon transport is calculated using the equation (6) defined as [1].

$$C_{Rn}\,(Bq\,m^{-3}) = \frac{C_{Ra} * \mathcal{E} * \rho_i * (1-\mathcal{E})}{\mathcal{E}} \quad (6)$$

Where ρ_i is the true density of soil (2.65 kg m^{-3}).

Radon surface exhalation rates FRs based on the fact that the surface is flat is calculated using the following equation (7) [7].

$$FR_s(Bq\,m^{-2}s^{-1}) = C_{Ra}\rho_b\mathcal{E}\left(\frac{T}{273}\right)^{0.75}\sqrt{\lambda D_0 P_t e^{(-6s)P_t - 6s^{14}P_t)}} \quad (7)$$

Where C_{Ra} is radium concentration, ρ_b is the bulk density, Ɛ is the emanation coefficient, T is the room temperature of dry soil, s is water saturation fraction, which is the ratio of volumetric water content to porosity P_t.

Radiation Indices

Human beings are exposed to γ-rays and α-particles, mainly from the ^{226}Ra, ^{232}Th and ^{40}K present in terrestrial materials. To assess the collective impact of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in a single quantity; radiation indices namely elemental concentration in ppm, radium equivalent activity, outdoor and indoor doses, annual effective dose and excessive lifetime cancer risk were calculated using procedures given in the literature.

The activity concentrations of ^{226}Ra in chain equilibrium with ^{238}U, ^{232}Th and ^{40}K in Bq kg^{-1} were converted into massive elemental concentrations in part per million ppm units for radium and thorium, and % by weight for potassium, respectively, according to the following equation (8) [17].

$$F_E = \frac{M_E W}{\lambda_{E,i} N_A f_{E,i}} C_{E,i} \quad (8)$$

where F_E is the fraction of element E in the sample, M_E is the atomic mass (kg mol^{-1}), $\lambda_{E,i}$ is the decay constant of the measured isotope of element E (s^{-1}), $f_{E,i}$ is the fractional atomic abundance in nature, and $C_{E,i}$ is the measured specific activity (Bq kg^{-1}) of the radionuclide under consideration ^{226}Ra, ^{232}Th and ^{40}K, N_A is the Avogadro's number 6.023×10^{23} atoms mol^{-1}, and W is a constant with value of 1,000,000 for U and Th concentration in ppm or 100 for K concentration in % of mass fraction.

For the purpose of comparing the radiological effect or activity of materials that contain ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them, a common index termed the radium equivalent activity Ra_{eq} is used. This activity index provides a useful guideline in regulating the safety standards on radiation protection for the general public residing in the area under investigation. The Ra_{eq} index represents a weighted sum of activities of the above mentioned natural radionuclides and is based on the estimation that 1 Bq kg^{-1} of ^{226}Ra, 0.7 Bq kg^{-1} of ^{232}Th and 13 Bq kg^{-1} of ^{40}K produce the same gamma radiation dose rates. The index is given as equation (9).

$$Ra_{eq} = C_{Ra} + (1.43 * C_{Th}) + (0.077 * C_K) \quad (9)$$

Where C_{Ra}, C_{Th} and C_K are the average activity concentration in the sample in Bq kg^{-1} of ^{226}Ra, ^{232}Th, and ^{40}K respectively [1].

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Excess ^{226}Ra which could come from the dumpsites soils is usually estimated according to the following equation (10) [18].

$$Ra_{fz}\,(Bq\,kg^{-1}) = C_{Ras} - C_{Ths}X\left(\frac{U}{Th_N}\right)XA \quad (10)$$

Where C_{Ras} and C_{Ths} are the activity concentrations of ^{226}Ra and ^{232}Th in the soil, $\frac{U}{Th_N}$ is the average value of the naturally observed $\frac{U}{Th_N}$ concentration ratio 0.23 and A is a conversion factor from ^{238}U concentration to ^{226}Ra radioactivity since 1mg kg^{-1} of ^{238}U is equal to 12.3 Bq kg^{-1} of ^{226}Ra when the two isotopes reached secular equilibrium.

In radiological health hazard assessment studies, activity utilization index AUI is usually calculated in order to estimate the dose rates in air from different combinations of the various radionuclides present in soils. It is given by the following equation (11) [19].

$$AUI = \left(\frac{C_{Ra}}{50\,Bq\,kg^{-1}}\right)F_{Ra} + \left(\frac{C_{Th}}{50\,Bq\,kg^{-1}}\right)F_{Th} + \left(\frac{C_K}{500Bq\,kg^{-1}}\right)F_K \quad (11)$$

Where C_{Ra}, C_{Th}, C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the dump site soil respectively, F_{Ra} (0.462), F_{Th} (0.604) and F_K (0.041) are the fractional contributions to the total dose rate in air due to gamma radiation.

The outdoor external dose D_{out} at 1m above the ground surface was assessed from the γ-radiation originating from ^{226}Ra, ^{232}Th and ^{40}K supposed to be equally distributed in ground. For the conversion of γ-radiation originating from ^{226}Ra, ^{232}Th and ^{40}K, the factors of 0.436, 0.599, 0.0417 nGy h^{-1} Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K were used for calculating the D_{out}. The D_{out} was calculated using the following equation (12) by European Commission, [20]:

$$D_{out}(nGy\,h^{-1}) = 0.436C_{Ra} + 0.599C_{Th} + 0.0417C_K \quad (12)$$

The γ -ray dose D_{in} imparted by ^{226}Ra, ^{232}Th and ^{40}K present in the indoor was calculated by converting the absorbed dose rate into effective dose using the three conversion factors; 0.92 nGy h^{-1} Bq kg^{-1} for ^{226}Ra, 1.1 nGy h^{-1} Bq kg^{-1} for ^{232}Th and 0.081 nGy h^{-1} Bq kg^{-1} for ^{40}K. By utilizing the above mentioned conversion factors, the following equation (13) was used to calculate the D_{in} [20].

$$D_{in}\,(nGy\,h^{-1}) = 0.92C_{Ra} + 1.1C_{Th} + 0.081C_K \quad (13)$$

Total gamma radioactivity can be used by health physicists especially those that are working in the University of Medical Sciences and Gani Fawehinmi Diagnostics Centre for acquiring information on the distribution of radiation exposures rate. Therefore, annual effective dose equivalent E to the population can be calculated using the conversion coefficient from absorbed dose in air to effective dose 0.7 Sv Gy^{-1}, the outdoor occupancy factor 0.2 and the indoor occupancy factor 0.8. Therefore, the annual effective doses outdoors and indoors equivalent were calculated by using the equations (14 and 15) [21].

$$E_{out}\,(mSv\,y^{-1}) = (D_{out}(nGy\,h^{-1}) X24hrX365.25daysX0.2X0.7\,Sv\,Gy^{-1})X10^{-6} \quad (14)$$

$$E_{in}\,(mSv\,y^{-1}) = (D_{in}(nGy\,h^{-1}) X24hrX365.25daysX0.8X0.7\,Sv\,Gy^{-1})X10^{-6} \quad (15)$$

The activity concentration in each of the samples is only an indication of the levels of radionuclides present. It does not relate the effect of such level on ecosystem. Therefore evaluation of its excessive lifetime cancer risk and other radiological hazard indices is important to ascertain the level of activity on human being. Excess Lifetime Cancer Risk *ELCR* was calculated using the below equations (16 and 17) for outdoor and indoor doses as:

$$ELCR_{outdoor} = E_{out} * LE * RF \quad (16)$$

$$ELCR_{in} = E_{in} * LE * RF \quad (17)$$

Where E_{out} and E_{in} are the annual effective doses, LE life expectancy (66 years) and RF (Sv^{-1}) is fatal risk factor per Sievert, which is 0.05 from ICRP-60 [22].

RESULTS

Radon

The mean value of R_M determined for the soil samples varied from S_3 (1.5 μBq kg^{-1} s^{-1}) to S_{16} (34.3 μBq kg^{-1} s^{-1}) with weighted mean of 15.2 μBq kg^{-1} s^{-1} as seen in the eleventh column of table 1. The mean value of C_{Rn} measured for the

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soil samples varied from S_3 (0.02 kBq m^{-3}) to S16 (0.49 kBq m^{-3}) with weighted mean of 0.22 kBq m^{-3} in the twelfth column of table 1. The weighted mean was within the permissible limit (0.2 kBq m^{-3}) recommended for exposure to radon by member of public [23]. Correlation between radium and radon emanation coefficient was carried out using Origin 8 statistical software. Figure 2 shows a strong correlation between ^{226}Ra and ^{222}Rn with correlation coefficient R^2=0.80981, which implies that ^{226}Ra and ^{222}Rn accompany with each other. The correlation was in good agreement with the value R^2=0.75 obtained in correlation between the radon exhalation rate and effective radium content in Iraq [24]. The mean value of radon surface exhalation rate FRs determined for the soil samples varied from S_3 (1.9 mBq m^{-1} s^{-1}) and S_{16} (42.3 mBq m^{-1} s^{-1}) with the weighted mean 18.8 mBq m^{-1} s^{-1}. However, Shigekazu *et al.* [25] revealed the results of previous studies in the earth's surface in some regions of the world as 8.0 (Northern part of North America), 19.5 (Southern part of North America), 23.3 (South America), 9.0 (Shout Asia), 17.3 (East Asia), 10.0 (Japan), 24.7 (Central Asia), 15.2 (South Asia), 17.8 (Europe), 29.1 (Africa), 33.4 (Australia), 11.5 (New Zealand) with the world's average 20.8 mBq m^{-1} s^{-1}.

Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the dumpsites varied from 20 ± 2 to 95 ± 14, 9 ± 1 to 49 ± 4 and 93 ± 35 to 615 ± 96 Bq kg^{-1} with average values of 58 ± 14, 23 ± 2 and 354 ± 53 Bq kg^{-1} respectively. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the control sites varied from 6 ± 1 to 89 ± 18, 6 ± 2 to 51 ± 14 and 78 ± 23 to 552 ± 66 Bq kg^{-1} with average values of 40 ± 14, 21 ± 2 and 206 ± 53 Bq kg^{-1} respectively. The profiles of the mean activity concentration of the various radionuclides with reference to sample numbers are shown in table 1. The total activity concentration ^{226}Ra + ^{232}Th + ^{40}K varied from 122 - 760 Bq kg^{-1} from S_2 down to S_{16} with average value 435 Bq kg^{-1}. In general, figure 3 shows that activity concentration of ^{40}K > ^{226}Ra > ^{232}Th. The average activity concentration of ^{226}Ra (58 ± 14 Bq kg^{-1}) in the soil samples was higher than world's average, while ^{40}K (354 ± 53 Bq kg^{-1}) and ^{232}Th (23 ± 2 Bq kg^{-1}) were lower than world's average that is 32, 412 and 45 Bq kg^{-1} respectively as per [32] world average values. The variations from the world's averages were attributed to different radioactive contents, emanation factors and diffusion coefficient of radon, as well as the porosity and density of the soil samples. The average of total activity concentration of ^{226}Ra + ^{232}Th + ^{40}K (435 Bq kg^{-1}) in the soil from the dumpsites and was also lower than the world's average of total activity concentration of these radionuclides in the soil that is 489 Bq kg^{-1}. Correlations between ^{40}K and ^{226}Ra, ^{232}Th and ^{40}K and ^{232}Th and ^{226}Ra are R^2=0.50578, 0.23644, -0.02249. The resultant correlation was moderately strong between ^{40}K and ^{226}Ra and weak between ^{232}Th and ^{40}K and ^{232}Th and ^{226}Ra respectively. The negative correlation between ^{232}Th and ^{226}Ra R^2=-0.02249 indicates high ^{226}Ra (^{238}U) enrichment. Weak correlation between ^{232}Th and ^{40}K R^2=0.23644 was due to the lower activity concentration of ^{232}Th compared with high concentrated ^{40}K. The moderately positive correlation between ^{40}K and ^{226}Ra R^2=0.50578 indicates the two radionuclides accompany each other. The $^{226}Ra/^{40}K$ and $^{232}Th/^{40}K$ ratios given in table 2 are relatively higher than the world's average of 0.067. These indicate that the study area is composed of rocks having low potassic values [33].

As reported, the elemental concentration of potassium, radium and thorium were calculated for the sixteen (16) sites considered in the studied area. The calculated elemental concentrations of thorium, radium and potassium are shown in table 1. The highest elemental concentrations of potassium were exhibited by samples at S_{15} and S_{16} with values that reach levels 1.8 and 2.0 % respectively. S_{10} and S_{16} have the highest concentrations of radium reaching levels of 7.6 and 7.7 ppm respectively. In addition, S_{12} and S_{14} present the highest concentration of thorium reaching 11.8 and 12.1. The lowest concentrations of thorium, radium and potassium were exhibited by S_2

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(2.2) ppm, S_3 (1.6) ppm and S_8 (0.3 %) respectively. As recorded S_{16} and S_{14} have the highest elemental concentrations, while S_8, S_3 and S_2 have the lowest elemental concentration for potassium, radium and thorium, respectively for all the radionuclides investigated. The measured weighted elemental mean in this study are 1.1 %, 4.8 ppm and 5.8 ppm. Using the weighted mean of all studied sites, the ratios of the elemental concentrations are $^{232}Th/^{226}Ra$=1.21ppm,$^{40}K/^{226}Ra$=0.23ppm and $^{40}K/^{232}Th$=0.19ppm. The ratio of weighted mean of $^{232}Th/^{226}Ra$=1.21ppm was much lower than the Clark's value 3.5, which indicates R-enrichment in the soil samples. The histograms and probabilities distributions of elemental concentration of ^{232}Th, ^{40}K and ^{226}Ra in soil samples are plotted using Origin 8.0 software and are shown in figure 4a-c. The presented graphs demonstrate that the considerable part of the data are contained within the range of 4-6 and 2-4 ppm for ^{232}Th content, 1.0-1.5 and 0.5-1.0 % for ^{40}K content and 7-8 and 4-5 for ^{226}Ra content respectively. The site with the highest number of radium content of 7.7 ppm is S_{16}. This result was predictable, since soils samples in the site contain the highest concentration value 95 ±14 Bq kg^{-1} of radium. However this fell within world's range from 0.1 to 20 ppm [1]. Weighted mean of thorium was lower than world average 7.4ppm by a factor of 1.3; radium was higher than the world average (2.8 ppm) by factor 1.7 while potassium was relatively equal to world average (1.3 %) in the upper crust of the Earth as seen in table 1.

The values of the Ra_{eq} calculated for the soil sample of Ondo city ranged from 56.00-200.91 Bq kg^{-1} with an average 119.11 Bq kg^{-1}. The average was 29 % higher than 34.98 Bq kg^{-1}[34] obtained in Uttar Pradesh Province in Northern India but less than 370 Bq kg^{-1} recommended as the world average and meets the recommended limit set by OECD [35]. Detail of Ra_{eq} and other radiation indices calculated during the present study are given in table 2.

As shown in table 2, the Ra_{fz} value ranged from -86.8 to 39.1 and has an average value -7.9Bq kg^{-1}. This shows that certain percentage of ^{226}Ra is leaked away and an indication that waste dumpsites are not well protected from rainfall and become hydraulically connected to surface water and shallow aquifers. The calculated values of the AUI in soil varied from 5.1 to 17.5 with mean value of 10.5. This indicates that the AUI mean (10.5) ≥ 2 corresponds to annual effective > 0.3 mSv y^{-1}. The outdoor external dose D_{out} due to the presence of ^{226}Ra, ^{232}Th and ^{40}K in the soil of Ondo city was calculated and it ranged from 23.32 to 91.64 nGy h^{-1} with an average value of 54.26 nGy h^{-1} as seen in table 2. The average was lower than the worlds' average of 59 nGy h^{-1} from UNSCEAR Report [36].

The value of D_{in} calculated during present study ranged from 30.71 to 182.30 with an average of 107.04 nGy h^{-1}, and was 1.27 times higher than the world's average of 84 nGy h^{-1}. The E_{out} and E_{in} calculated for the soils were given in table 2. E_{out} ranged from 0.03 to 0.11 with an average of 0.07 mSv y^{-1} which was less than the world's average of 0.41 mSv y^{-1} [1], while E_{in} ranged from 0.15 to 0.89 with an average 0.52 mSv y^{-1} which was higher than the world's average 0.41 mSv y^{-1} [1]. The total average annual effective dose E_{in} + E_{out} was estimated to be (0.07 + 0.52) 0.59 mSv y^{-1} and was 1.13 times higher than the world's average of 0.52 mSv y^{-1} but lower than the criterion limit of 1 mSv y^{-1} considered from ICRP-60. The total average annual effective 0.59 mSv y^{-1} showed agreement with AUI mean value 10.5≥2 that gives effective dose equivalent >0.3 mSv y^{-1}. The calculated outdoor ELCR outdoor ranged from 0.09 × 10^{-3} to 0.37 × 10^{-3} with an average value of 0.22 x 10^{-3} and for indoor exposure it was from 0.50 × 10^{-3} to 2.93 × 10^{-3} with an average of 1.73 × 10^{-3}. The total ELCR ranged from 0.59 × 10^{-3} to 3.33 × 10^{-3} with an average value of 1.95 × 10^{-3} as shown in table 2. The total *ELCR* was 1.34 times higher than the world's average 1.45 × 10^{-3}, 2.1 times 0.95 × 10^{-3}, 89 % of 2.17 × 10^{-3} and 61 % of 3.21 × 10^{-3} obtained from Tulkarem Palestine, Azad Pakistan and Northern Pakistan respectively [33, 37, 38]. The total *ELCR* indicates that people from the study falls in the category of moderate activity concentration.

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Table 1. Mean Activity levels of ^{40}K, ^{226}Ra and ^{232}Th (Bq kg-^{1}) in the Samples, Elemental Concentration (ppm and% for ^{40}K) and Radon Parameters (% , Bq kg^{-1} s^{-1}x10^{-6}, kBq m^{-3}, mBq m^{-1}s^{-1}).

Zone	Site	No of Sample	Activity Concentration			Elemental concentration			Radon Parameters			
			^{40}K	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	ε	R_M	C_{Rn}	ERs
			Mean			Mean			Mean			
1	S1	4	291±97	31±3	17±2	0.9	2.5	4.2	5.6	3.6	0.14	4.5
	S2	5	313±4	40±5	9±1	1.0	3.2	2.2	6.0	5.0	0.07	6.2
	S3	4	134±48	20±2	18±2	0.4	1.6	4.4	3.6	1.5	0.02	1.9
	S4	5	420±36	85±9	21±2	1.4	6.9	5.2	14.0	25.0	0.36	30.8
2	S5	3	455±47	73±22	19±2	1.5	5.9	4.7	13.2	20.2	0.29	25.0
	S6	4	235±78	53±17	15±1	0.8	4.3	3.7	9.6	10.7	0.15	13.2
	S7	4	434±40	90±27	18±2	1.4	7.3	4.4	20.0	37.8	0.54	46.6
	S8	3	93±35	24±2	15±1	0.3	1.9	3.7	4.3	2.2	0.03	2.7
3	S9	5	193±68	61±19	11±1	0.6	4.9	2.7	8.0	10.2	0.15	13.0
	S10	4	384±19	94±28	21±2	1.2	7.6	5.2	13.0	25.7	0.37	32.0
	S11	4	409±33	35±12	21±2	1.3	4.2	5.2	6.3	4.7	0.07	5.7
	S12	4	335±9	49±16	48±4	1.1	4.0	11.8	8.9	9.1	0.13	11.3
4	S13	4	268±89	43±15	26±3	0.9	3.5	6.4	7.8	7.0	0.10	8.7
	S14	4	510±64	56±17	49±4	1.7	4.5	12.1	15.0	17.6	0.25	21.8
	S15	5	568±82	86±17	26±4	1.8	7.0	6.4	15.5	28.6	0.40	34.5
	S16	3	615±96	95±14	41±4	2.0	7.7	10.1	17.2	34.3	0.49	42.3
Weighted Mean			354±53	58±14	23±2	1.1	4.8	5.8	10.5	15.2	0.20	18.8

Comparison of the Activity and Elemental Concentrations with other countries of the world

No.	Country	Activity Concentration			Total	References	Country	Elemental Concentration			References
		^{40}K	^{226}Ra	^{232}Th				^{40}K	^{226}Ra	^{232}Th	
1	Nigeria	354	58	23	435	Present study	Nigeria	1.1	4.8	5.8	Present Study
2	Tamilnadu, India	401	5	34	540	(8)	Rajasthan India	0.2-0.5	2.6-6.3	10.6-26.1	(14)
3	India Palar	472	10	36	645	(26)	Cyprus	<1-1.9	<1-3.2	<1-9.8	(29)
4	Turkey, Maritza	472	64	36	572	(27)	Cyprus	0.6	0.9	2.8	(30)
5	South Carolina	609	21	45	665	(28)	Istanbul Turkey	1.1	1.7	9.1	(31)

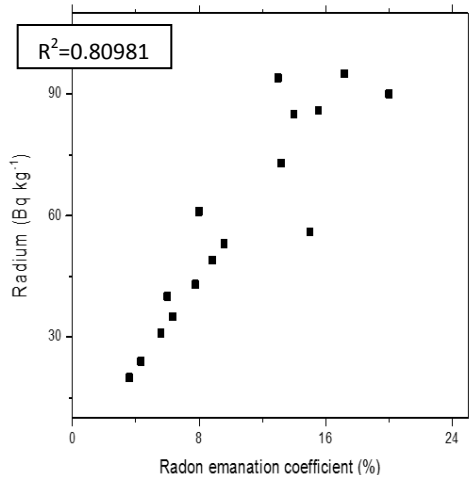

Figure 2. Correlation between ^{226}Ra and ^{222}Rn.

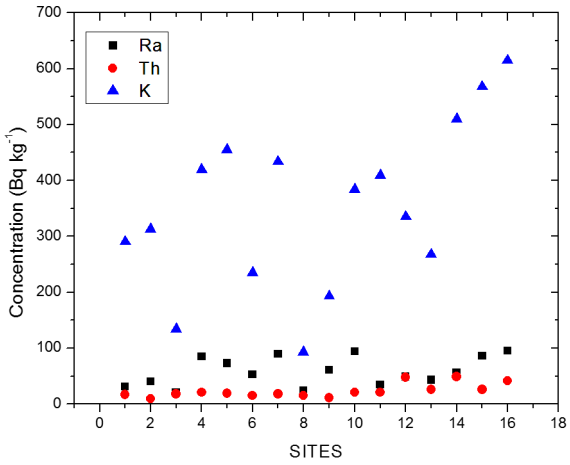

Figure 3. Concentration of ^{226}Ra, ^{232}Th and ^{40}K.

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Figure 1. Modified Ondo Google Satellite Map Showing Zones of Sample Collection. Map data ©2017 Google (14)

Radionuclides	Minimum	Maximum	Average	St deviation	World Average
^{226}Ra (Bq kg^{-1})	20±2	95±14	58±14	25.76	32[32]
^{232}Th (Bq kg^{-1})	9±1	49±4	23±2	12.17	45[32]
^{40}K(Bq kg^{-1})	93±35	615±96	354±53	149.03	412[32]
^{226}Ra+^{232}Th+^{40}K(Bq kg^{-1})	122±38	760±14	435±69	175.31	489[32]
^{226}Ra/^{40}K	0.09	0.32	0.17	0.07	0.067[32]
^{232}Th/^{40}K	0.04	0.16	0.07	0.04	0.067[32]
^{226}Ra/^{232}Th	1.11	5.55	2.91	1.53	1.00[32]
Radiation Indices					
Ra_{eq}(Bq kg^{-1})	56.00	200.91	119.11	43.98	370[1]
Ra_{fz} (Bq kg^{-1})	-86.84	39.06	-7.89	38.34	-
AUI	5.05	17.51	10.48	3.88	-
D_{out} (nGyh^{-1})	23.32	91.64	54.26	20.10	59[1]
D_{in} (nGyh^{-1})	30.71	182.30	107.04	42.20	84[1]
E_{out} (mSv y^{-1})	0.03	0.11	0.07	0.02	0.07[1]
E_{in} (mSv y^{-1})	0.15	0.89	0.52	0.21	0.41[1]
E_{out}+E_{in}(mSv y^{-1})	0.18	1.00	0.59	0.23	0.52[33]
ELCRx10^{-3} (out)	0.09	0.37	0.22	0.08	0.29[33]
ELCR x 10^{-3} (in)	0.50	2.93	1.73	0.68	1.16
ELCR x10^{-3} (Total)	0.59	3.33	1.95	0.76	1.45

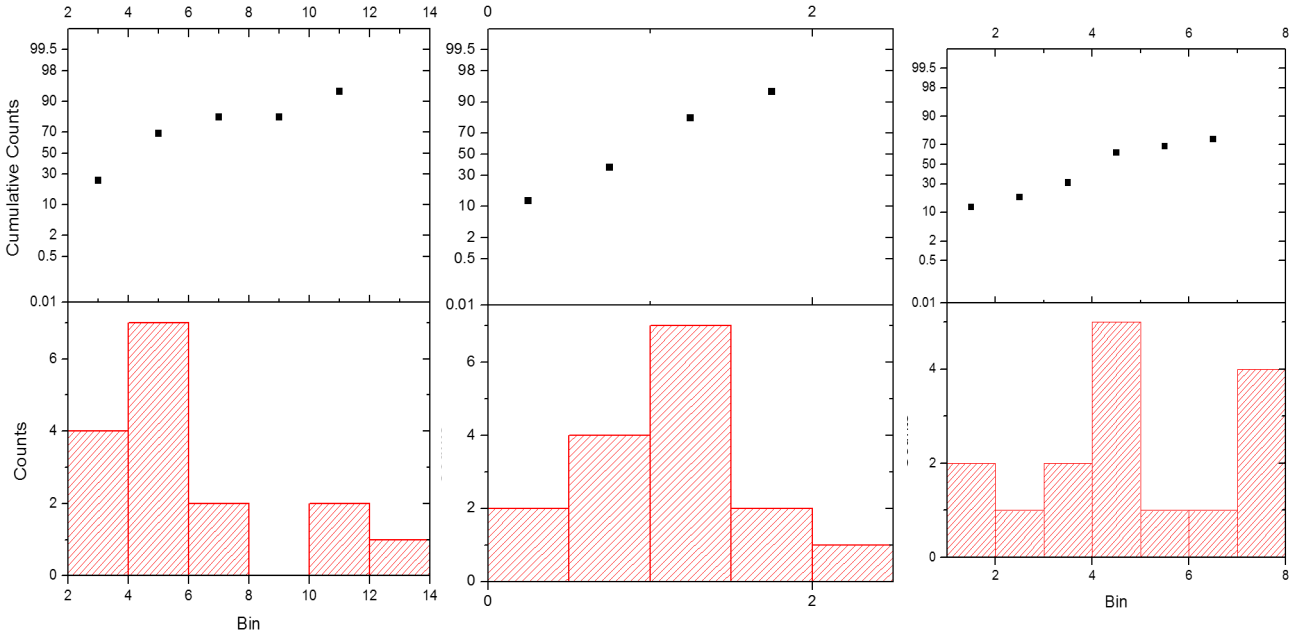

Figure 4. a. Frequency distribution of elemental concentration of ^{23}2Th. **b.** Frequency distribution of elemental concentration of ^{40}K. **c.** Frequency distribution of elemental concentration of ^{226}Ra.

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DISCUSSION

The present study with radon weighted mean 18.8 mBq m^{-1} s^{-1} fell within the global averages 21 mBq m^{-1} s^{-1} (1 atom cm^{-2} s^{-1}), 26 mBq m^{-1} s^{-1}, 33 mBq m^{-1} s^{-1} [25] and predicted global average 34 mBq m^{-1} s^{-1}[25]. It implies no significant radiological hazard for human population living in the area. The background range of natural radioactivity in the soil of the studied city was the same as the range of typical values recorded in the upper part of the earth's crust. It can be rightly said that the soil from the waste dumpsites in Ondo exhibit natural activities. The $^{226}Ra/^{232}Th$ ratio was higher than the world's average of 1; this revealed the contribution of wastes to background radionuclides. Comparison of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the dumpsites with some other countries of the world are presented in table 1. This showed that the studied area fell within acceptable limit and natural radioactivity majorly depends on the human activities, geological and geographical conditions of the studied area. A comparison of radionuclides and radiological hazard indices were given in table 2. From the table 2 it can be seen that values of radionuclides and radiation hazard indices such as Ra_{eq} and D_{out} of soil from the waste dumpsites of Ondo estimated during present study were slightly lower than world average while parameters such as D_{in}, E_{in}, E_{out}+E_{in} were slightly higher than the world's average for most of the radiological indices. These indicate the contribution of waste to background radionuclides in the soil.

CONCLUSION

The natural activities concentration of ^{40}K, ^{226}Ra and ^{232}Th in soil from wastes dump sites within Ondo metropolis had been determined. The average radium equivalent activity, outdoor external dose and outdoor annual effective dose were lower than the world's average. The indoor internal dose, indoor annual effective dose and total annual effective dose were slightly higher than the world's average limits. The ELCR factor assessed during present study on the basis of outdoor E_{out} and indoor annual dose E_{in} was slightly higher than world's average. Although the city is safe, but no matter how small radiation exposure or dose may be it has effect on human beings and the reported values may indicate no immediate health hazards, but may cause long-term health hazard to the Dumpsites workers, scavengers and residents of the host communities. Therefore, it is advised that there should be a regular monitoring/inspection of radiation levels in the environments by the government and people. Residents should start to reduce solid waste production by concentrating on a few strategies such as purchasing products with less packaging, purchasing products in bulk or larger sizes, purchasing more durable products maintaining properly and repairing instead of replacing.

***Conflicts of interest*:** *Declared none.*

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