Activity Concentration Measurement of Naturally-Occurring Radionuclides in Various Vegetation plots in Rustenburg, South Africa

P O Olagbaju[1*], O B Wojuola[1] and M V Tshivhase[2]

[1] Department of Physics, North-West University, Mafikeng Campus, Mmabatho, South Africa
[2] Centre for Applied Radiation Science and Technology, North-West University, Mafikeng Campus, Mmabatho, South Africa

E-mail: olagbajupetero@gmail.com

Abstract. Globally, radiation level varies from one region to another due to the differences in the geological and mineralogical composition, and on the industrial and agricultural activities in each region. The aim of this study is to assess soil radioactivity level in different vegetational plots in Rustenburg which is associated with mining, industrial and agricultural practices in the North-West province of South Africa. In this study, the activity concentration of naturally-occurring radionuclides in beetroots (BRS), leeks (LKS), mints (MTS), onion (ONS), parsley (PSS) and wheat (WTS) plots were measured using broad-energy germanium (BEGe) detector, and correlation matrix was used to study the relationship between the radiological level in all the vegetation plots studied. The mean activity concentration of (^{238}U, ^{232}Th, and ^{40}K) was observed to be (25.15 ±1.14 Bq/kg, 21.04±9.49 Bq/kg and 90.20±3.76 Bq/kg), (11.48±0.68 Bq/kg, 6.77±0.18 Bq/kg and 51.30±4.96 Bq/kg), (23.63±1.35 Bq/kg, 15.45±0.28 Bq/kg and 105.10±7.74 Bq/kg), (11.29±0.76 Bq/kg, 8.08±0.19 Bq/kg and 45.26±13.78 Bq/kg), (23.08±1.50 Bq/kg, 19.52±0.30 Bq/kg and 99.69±6.19 Bq/kg) and (11.78±0.75 Bq/kg, 8.32±0.38 Bq/kg and 89.25±11.86 Bq/kg) for soil collected in BRS, LKS, MTS, ONS, PSS and WTS plots respectively. These were observed to be lower than the world average values of 30 Bq/kg, 35 Bq/kg and 400 Bq/kg reported by United Nation Scientific Committee on the Effects of Atomic Radiation [1], for ^{238}U, ^{232}Th and ^{40}K respectively. Radium equivalent activity, which is the weighted sum of the activity concentration of ^{238}U, ^{232}Th, and ^{40}K in measured soil samples, was found to be below the world's average value of 370 Bq/Kg, which show that the study area is safe for living and agriculture purposes. Weak correlation of radium equivalent activities was observed only in beetroot-wheat and leeks-parsley. This suggests different influence of plant types on soil radionuclides and thus affect their choice for phytoremediation purpose.

1. Introduction

Humans are continuously exposed to ionizing radiation from natural and anthropogenic sources. Naturally-occurring radionuclides originate from rock and mineral weathering as a result of volcanic eruption, erosion [1]. They contribute about 96% of the total radiation exposure to humans [1, 2]. Anthropogenic activities such as mining, industrial, and agricultural practices have been reported to enhance the concentration of naturally-occurring radionuclides in the soil. Their migration into non-contaminated areas via erosion, atmospheric deposition, penetration into environmental media pose significant radiological risk to human health [3].

Soil serves as a direct source of radionuclides, leading to the contamination of agricultural products, which represent an ingestion pathway of radionuclides to man [4, 5]. Soil radioactivity varies from one region to another depending on the geological and anthropogenic condition of each region, and is also influenced by the parent rock and its formation process [6, 7]. Since soil serves as indicator to environmental radiological contamination, the knowledge of soil radioactivity level is essential in environmental monitoring and protection, as it can be used to predict changes in environmental radioactivity caused by human activities [1].

South Africa, which has one of the most diverse and comprehensive crop farming systems, is long associated with mining and industrial activities resulting in radioactive waste littered in farmlands and communities [8]. Mine tailings and industrial wastes are major source of contamination in the environment, and their accumulation in soil, air, water, and agricultural products via the atmospheric deposition, groundwater sources and surface water body, represent a direct and indirect exposure pathway for incorporating into the human food chain. Therefore, this study aims to measure the activity concentrations of natural radionuclides (^{238}U, ^{232}Th, and ^{40}K) in different vegetational plots, and to investigate the relationship between various plots and soil radioactivity.

2. Materials and Methods

2.1. Sample collection, preparation, and measurement

Soil samples were randomly collected from seven vegetational plots. At each sampling point, four soil samples were collected with a hand trowel, mixed to form a homogenous composite sample. Soil samples were collected at a depth of about 20 cm to avoid the effect of plant cycling of radionuclides. They were transferred to the laboratory in well-labelled polythene bags, where they were processed for measurement, as described by [9, 10]. Collected soil samples were air-dried to remove moisture contents, crushed into fine powered using an electric grinder, and sieved using a 2 mm mesh screen to obtain a fine-grained homogenous soil sample. The fine-grained homogeneous sample was packed and sealed into well-labelled airtight containers to prevent the escape of ^{220}Rn and ^{222}Rn. The sealed soil samples were stored for about four weeks to allow secular radioactive equilibrium between thorium, radium and short-lived decay products before measurement at the Centre for Applied Radiation Science and Technology (CARST), North-West University South Africa [4].

Activity concentration of naturally-occurring radionuclides in soil was measured using broad-energy germanium detector (BEGe), with relative efficiency of 60%, a resolution of 2.0 keV for 1332 keV gamma-ray emission of ^{60}Co and necessary efficiency and energy calibration. Each sample was counted for 43,200 seconds, for the determination of the activity concentrations of ^{238}U using the 295.2 keV (19.7%) and 351.9 keV (38.9%) gamma-rays from ^{214}Pb and the 609.3 keV (43.3%), 1120.3 keV (15.7%) and 1764.5 keV (15.1%) gamma-rays from ^{214}Bi, and activity concentration of ^{232}Th using 238.6 keV (44.6%) from ^{212}Pb and 338.3 keV (11.4%), 911.6 keV (27.7%) and 969.1 keV (16.6%) gamma ray of ^{228}Ac, while the activity concentration of ^{40}K was determined using its 1460 keV gamma-line [11].

The activity concentrations $C(Bq/kg)$ of the radionuclides were calculated using equation (1), as given by[10, 11]:

$$C(Bq/kg) = KC_n \tag{1}$$

where Cn is the count rate under the corresponding peak, $K = \frac{1}{\varepsilon \rho_\gamma M_s}$ is the efficiency of the detector at specific gamma-ray energy, ρ_γ is the absolute transition probability of the specific gamma-ray, M_s is the sample mass. The minimum detectable activity of ^{40}K was 0.0189 Bq, while ^{238}U and ^{232}Th gamma energy line were not detected in the background spectrum.

2.2. Radiation indices measurements

Absorbed dose rate (ABDR) in air at 1 m from terrestrial sources of gamma radiation is estimated using the measured activity concentration of ^{238}U, ^{232}Th and ^{40}K by applying concentration-to-dose conversion factors of 0.462, 0.604 and 0.0417 respectively [1]

$$ABDR\ (nGy/hr) = 0.462C_U + 0.604C_{Th} + 0.0417C_K \qquad (2)$$

Annual effective dose equivalent was estimated using the conversion coefficient of 0.70 Sv/Gy from absorbed dose in air to effective dose received by adults and 0.2 as the outdoor occupancy factor and 8760 hr [11]

$$AEDE\ (mSv/yr) = ABDR\ (nGy/hr) \times 8760 \times 0.2 \times 0.70 \times 10^{-6} \qquad (3)$$

The annual gonadal equivalent dose (AGED) equivalent due to specific activities of ^{238}U, ^{232}Th, and ^{40}K was estimated using the formula given by [12]

$$AGED\ (\mu Sv/yr) = 309C_U + 4.18C_{Th} + 0.314C_K \qquad (4)$$

Excess lifetime cancer risk (ELCR), used to estimate the probability of developing cancer over a lifetime at a given exposure level [12] is calculated using:

$$ELCR = AEDE\ (mSv/yr) \times D_L(yr) \times RF(Sv^{-1}) \times 10^{-3} \qquad (5)$$

where, AEDE is the annual effective dose equivalent, D_L is the average duration of life (estimated to be 70 years), RF is the Risk Factor (Sv^{-1}) which is given as 0.05 for stochastic by ICRP [13].
External hazard index is used to quantify the effect of radon, a progeny of radium, and its short-lived products to the respiratory organs. It is estimated using equation 6, and must be less than unity for the radiation hazard to be negligible [12].

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \qquad (6)$$

3. Results and Discussion

The activity concentration of naturally-occurring radionuclides in sampled plots, which include beetroot plot (BRS), leek plot (LKS), mint plot (MTS), onion plot (ONS), parsley plot (PSS) and wheat plot (WHTS) are presented in table 1. The results obtained for activity concentration of naturally-occurring radionuclides in all the measured plots were observed to be lower than the world average recommended values of 30 Bq/kg, 35 Bq/kg and 400 Bq/kg for ^{238}U, ^{232}Th and ^{40}K, respectively [1]. The prevailing activity concentration of potassium in all sample soils can be attributed to its abundance in nature, agricultural practices that involve the use of organic and inorganic fertilizer containing potassium [14]. The high concentration of ^{238}U to ^{232}Th is attributed to the presence of uranium-bearing minerals associated with mining regions, which is predominant in the study area [15]. The variation observed in the activity concentration of each radionuclide is attributed to differences in radionuclides solubility and mobility [16].

The relationship between radium equivalent activity, which is used to express the weighted sum of ^{226}Ra (^{238}U), ^{232}Th, and ^{40}K, was investigated for the sampled plots using Pearson correlation. From table 2, a weak positive correlation can be observed between radium equivalent activity in beetroot-wheat, and leeks-parsley. Pearson correlation of radium equivalent activity among other vegetational plots show poor correlation coefficients. This observation suggests the varying influence of plant types on soil radionuclides, affecting choice of plant for phytoremediation purpose.

Table 1: Activity concentration of naturally-occurring radionuclides of soil in the selected area.

Sample code	Parameters	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Ra_{eq} (Bq/kg)
	Maximum	29.56±1.19	23.23±0.33	112.70±4.69	69.46
BRS	Minimum	20.71±1.25	19.10±0.31	52.48±3.76	55.72
	Average	25.15±1.14	21.04±9.49	90.20±5.94	62.18±3.95
	Maximum	14.75±0.67	8.154±0.20	68.08±3.30	30.55
LKS	Minimum	8.87±0.63	5.37±0.22	34.52±3.43	21.04
	Average	11.48±0.68	6.77±0.18	51.30±4.96	24.92±3.36
	Maximum	30.99±1.44	20.94±0.34	149.10±4.98	66.93
MTS	Minimum	12.75±0.24	12.35±0.24	57.98±6.38	45.09
	Average	23.63±1.35	15.45±0.28	105.10±7.74	53.81±6.50
	Maximum	18.35±0.97	10.53±0.21	97.03±4.61	35.95
ONS	Minimum	6.06±0.59	6.79±0.17	45.26±13.78	22.64
	Average	11.29±0.76	8.08±0.19	70.42±10.06	28.26±4.78
	Maximum	25.35±1.31	18.38±0.28	133.50±2.60	50.79
CMR	Minimum	10.78±0.69	10.16±0.22	26.48±4.46	32.66
	Average	16.14±0.85	13.68±0.25	70.52±6.56	41.13±5.45
	Maximum	26.79±0.89	21.87±0.31	134.40±5.82	64.09
PSS	Minimum	17.91±5.14	13.84±0.25	58.46±4.96	52.53
	Average	23.08±1.50	19.52±0.30	99.69±6.19	58.67±3.45
	Maximum	18.78±0.85	10.02±0.71	128.90±3.33	43.04
WHTS	Minimum	7.13±0.59	7.08±0.18	62.79±10.40	23.26
	Average	11.78±0.75	8.32±0.38	89.25±11.86	30.55±6.16
UNSCEAR [1]		35	30	400	370

Table 2: Correlation coefficient of radium equivalent in vegetational plots.

	BRTS	LKS	MTS	PSS	ONS	WHTS
BRTS	1.00					
LKS	-0.47	1.00				
MTS	-0.81	0.15	1.00			
PSS	-0.35	0.58	0.28	1.00		
ONS	-0.43	0.23	-0.10	-0.07	1.00	
WHTS	0.66	-0.54	-0.57	-0.07	0.02	1.00

3.1. Radiological hazards

The average absorbed dose rates estimated from the activity concentration of ^{238}U, ^{232}Th and ^{40}K, for beetroot, leeks, mints, onion, parsley and the uncultivated plot is 28.11 nGy/hr, 11.46 nGy/hr, 24.66 nGy/hr, 13.05 nGy/hr, 18.68 nGy/hr, 26.64 nGy/hr and 14.22 nGy/hr respectively. Absorbed dose rate obtained for all vegetational plots is found to be lower than the world's average value of 60 nGy/hr recommended by united Nations Scientific Committee on the Effect of Atomic Radiation [1] . Table 3 show the average annual effective dose equivalent (AEDE), excess lifetime cancer risk (ELCR), annual gonadal dose equivalent (AGDE) and external hazard due to gamma ray emission from naturally-occurring radionuclide from all investigated plots. Similarly, they are observed to be below the world average value of 1 mSv/yr, 2.90×10^{-04}, 300 µSv/yr and unity, for annual effective dose equivalent, excess lifetime cancer risk annual gonadal dose equivalent and external hazard index respectively, indicating that the study area is safe for living and agricultural purposes.

Table 3: Average radiological dose and hazard indices of soil in various vegetational plots.

Plots	ABDR (nGy/hr)	AEDE (mSv/yr)	ELCR	Hex	AGED (µSv/yr)
BRS	28.11	0.03	1.21×10^{-4}	0.17	122.78
LKS	11.46	0.01	4.91×10^{-5}	0.07	67.87
MTS	24.66	0.03	1.06×10^{-4}	0.15	122.74
ONS	13.05	0.02	5.59×10^{-5}	0.08	73.71
CMR	18.68	0.02	8.01×10^{-5}	0.11	88.75
PSS	26.64	0.03	1.14×10^{-4}	0.16	119.34
WHTS	14.22	0.02	6.08×10^{-5}	0.08	81.15
UNSCEAR [1]	60.00	1.00	2.90×10^{-04}	1.0	300.00

4. Conclusion

Agricultural practices play an essential role in environmental radioactivity level, posing a significant exposure scenario to farmers and human food chains. The radiological impact of agrochemicals in soil was estimated using soil radioactivity measured in different vegetational plots in Rustenburg, South Africa. In all sampled soil, the activity concentration of naturally-occurring radionuclides was observed to increase in order of $^{40}K > {}^{238}U > {}^{232}Th$. The prevailing activity concentration of potassium in all sampled soil is attributed to its abundance in nature, agricultural practices that involve the use of organic and inorganic fertilizer containing potassium. Observed high concentration of ^{238}U to ^{232}Th is has a result of the presence of uranium-bearing minerals associated with mining regions. The overall variation observed in the concentration of measured radionuclides is attributed to differences in their solubility and mobility [17].

However, radiological parameters such as absorbed dose rate, annual effective dose equivalent, excess lifetime cancer risk, external hazard index, estimated are found to be below recommended level. Thus, the study indicates the study area is safe for living and agricultural purposes. Notwithstanding, regular monitoring of soil radioactivity is recommended due to the presence of mines in the farming vicinity and the use of various agrochemicals, which contribute significantly to soil radioactivity.

References

[1] UNSCEAR. Sources and effects of ionizing radiation. Journal of Radiological Protection. 2000;21(1):83.

[2] Akpan AE, Ebong ED, Ekwok SE, Eyo JO. Assessment of radionuclide distribution and associated radiological hazards for soils and beach sediments of Akwa Ibom Coastline, southern Nigeria. Arabian Journal of Geosciences. 2020;13(15):1-12.

[3] Andersson KG. Migration of radionuclides on outdoor surfaces. Radioactivity in the Environment. 2009;15:107-46.

[4] IAEA-TECDOC-566. The Use of Gamma Ray Data to Define the Natural Radiation Environment. International Atomic Energy Agency Vienna; 1990.

[5] Ilori AO, Chetty N. Soil-to-crop transfer of natural radionuclides in farm soil of South Africa. Environmental Monitoring and Assessment. 2020;192(12):1-13.

[6] Gad A, Saleh A, Khalifa M. Assessment of natural radionuclides and related occupational risk in agricultural soil, southeastern Nile Delta, Egypt. Arabian Journal of Geosciences. 2019;12(6):1-15.

[7] Guidotti L, Carini F, Rossi R, Gatti M, Cenci RM, Beone GM. Gamma-spectrometric measurement of radioactivity in agricultural soils of the Lombardia region, northern Italy. Journal of environmental radioactivity. 2015;142:36-44.

[8] Kamunda C, Mathuthu M, Madhuku M. An assessment of radiological hazards from gold mine tailings in the province of Gauteng in South Africa. International Journal of environmental research and public health. 2016;13(1):138.

[9] Ogundele LT, Oluwajana OA, Ogunyele AC, Inuyomi SO. Heavy metals, radionuclides activity and mineralogy of soil samples from an artisanal gold mining site in Ile-Ife, Nigeria: implications on human and environmental health. Environmental Earth Sciences. 2021;80(5):1-15.

[10] Olagbaju P, Okeyode I, Alatise O, Bada B. Background radiation level measurement using hand held dosimeter and gamma spectrometry in Ijebu-Ife, Ogun State Nigeria. International Journal of Radiation Research. 2021;19(3):591-8.

[11] Ibikunle SB, Arogunjo AM, Ajayi OS. Characterization of radiation dose and soil-to-plant transfer factor of natural radionuclides in some cities from South-Western Nigeria and its effect on man. Scientific African. 2019;3:e00062.

[12] Avwiri G, Osimobi J, Agbalagba E. Natural Occurring Radionuclide Variation with a Soil Depth Profile of Udi and Ezeagu Local Government Areas of Enugu State, Nigeria. Facta Universitatis-Series: Working and Living Enviromental Protection. 2013;10(1):53-60.

[13] Taskin H, Karavus M, Ay P, Topuzoglu A, Hidiroglu S, Karahan G. Radionuclide Concentrations in Soil and Lifetime Cancer Risk due to Gamma Radioactivity in Kirklareli, Turkey. Journal of Environmental Radioactivity. 2009;100(1):49-53.

[14] Ahmed NK, El-Arabi AGM. Natural radioactivity in farm soil and phosphate fertilizer and its environmental implications in Qena governorate, Upper Egypt. Journal of Environmental Radioactivity. 2005;84(1):51-64.

[15] Ugbede FO, Osahon OD, Akpolile AF. Natural radioactivity levels of 238U, 232Th and 40K and radiological risk assessment in paddy soil of Ezillo rice fields in Ebonyi State, Nigeria. Environmental Forensics. 2021:1-16.

[16] Absar N, Abedin J, Rahman MM, Miah M, Hossain M, Siddique N, et al. Radionuclides Transfer from Soil to Tea Leaves and Estimation of Committed Effective Dose to the Bangladesh Populace. Life. 2021;11(4):282.

[17] Kritsananuwat R, Sahoo S, Arae H, Fukushi M. Distribution of 238 U and 232 Th in selected soil and plant samples as well as soil to plant transfer factors around Southern Thailand. Journal of Radioanalytical and Nuclear Chemistry. 2015;303(3):2571-7.

 ISBN: 978-0-620-97693-0