

ASSESSMENT OF CONCENTRATIONS AND DISTRIBUTIONS OF RADIONUCLIDES IN SOILS OF ABANDONED MINE SITES OF JOS SOUTH AND BARKIN LADI IN PLATEAU STATE NIGERIA.

ABSTRACT

Barkin Ladi and Jos South Local Government Area of Plateau State is situated in the central part of Northern Nigeria, on a rugged terrain of lowlands. It is the principal centre of tin and columbite mineralization, which forms the focal area of younger granites. The work was aimed at assessing the concentration and distribution of radionuclides in soils of abandoned tin mines and the soil's physicochemical properties. The samples were collected in July, from five different locations in both Barkin Ladi and Jos South where mining activities were predominant. The samples were analyzed using a gamma-ray spectrometer. The result obtained reveals a range of the concentrations of ^{40}K as 116.76 ± 0.21 to 250.15 ± 0.42 Bq/kg, ^{226}Ra from 24.37 ± 0.31 to 90.45 ± 0.86 Bq/kg, and ^{232}Th from 15.74 ± 0.71 to 88.43 ± 0.14 Bq/kg. The result of the physicochemical properties of the soil analyzed ranges from pH (3.12 to 6.81); organic carbon (0.01 to 1.62%); organic matter (0.22 to 3.01 %), cation exchange capacity (1.01 to 4.21Cmol/kg); clay (9.72 to 40.82 %); silt (8.32 to 40.32% and sand (32.42 to 75.38%) respectively. All the ^{40}K results analyzed in all the sampling sites were lower than the recommended limits of 412Bq/kg, whereas most of the results of ^{226}Ra and ^{232}Th were above the 33Bq/kg and 45Bq/kg permissible limits. The physicochemical properties of the soil varied across the sampling sites. Most of the sampling sites are polluted with radionuclides and migrations of the radionuclides from the ex-mining sites to other parts of the environment were noticed from the studies conducted.

Keywords: Mining point; Pollution; Radionuclides; Soil, and Tailings

INTRODUCTION

1.1 Background of the Study

The **geochemical behaviour** of thorium, uranium, and, to a lesser degree, rare earth elements (REEs) is comparatively comparable to that of other elements in the geological environment. The majority of environmental matrices contain radioactive elements including ^{232}Th and ^{238}U , as well as their decay products like ^{226}Ra . These elements can enter living things through a variety of paths, which can lead to the sources of human exposure [1].

Ever since life originally began in the universe, naturally occurring radioactive elements have exposed species including plants and animals to radiation [2]. There are more than 60 radionuclides in nature because of how radioactive our planet is [3]. The sources of radioactivity and the emitters of nuclear radiation that permeate our daily lives are these radionuclides. Everywhere there is radiation, humans are continually exposed to it, either directly or indirectly. Humans are exposed to radiation from a

wide range of natural and artificial sources. Ionizing radiation comes from a variety of sources, including mining sites, water, air, soils, and construction materials [3]. We breathe in or absorb radionuclides from the food we eat, and in the water we drink.

Populations in communities may be exposed to radiation from both natural and man-made sources. Both terrestrial and extraterrestrial (space) sources can contribute to the natural radiation that surrounds life on Earth. Ionizing radiation from radionuclides in the earth, rocks, building materials, water, air, and mining sites is referred to as global radiation. Conversely, cosmic rays are high-energy radiation from space that reaches the Earth's atmosphere [4]. Radiation-sensitive elements like ^{238}U , ^{232}Th , and the daughter products ^{226}Ra , ^{222}Rn , ^{235}U , and ^{40}K are significant naturally occurring radiation sources. Ionizing radiation, including α , β , and γ radiations, is released from many terrestrial materials. Among them, soil is a primary source of naturally occurring radioactivity and the primary pathway for radionuclide migration and transfer into the surrounding environment [6].

Numerous investigations have been conducted globally to ascertain the activity content of these radioactive elements, which are the primary causes of natural radiation in soil that humans have contaminated [7]. Geographical location and some human activities influence the extent of these natural exposures [8]. In addition to radiation from natural sources, radiation from man-made sources can also affect humans. Radiation applications may be found in many fields, including biology, agriculture, industry, medicine, and the production of electric power. Humans may come into contact with radiation from various radioactive sources during their uses, as well as radiation-related illnesses [9]. The study, therefore, is to determine the concentration and distribution (both spatial and temporal) of radionuclides in soils of abandoned tin mines in some parts of Barkin Ladi and Jos South Local Government Area of Plateau State.

MATERIALS AND METHODS

2.1 Materials

2.1.1 Reagent/ Solvents

The reagents that were used for the research work are 1N Potassium dichromate ($K_2Cr_2O_7$), sodium fluoride, 0.5N ferrous ammonium sulfate $Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$, Diphenylamine, Tetraoxosulphate (VI) acid 0.1M $BaCl_2 \cdot 2H_2O$, and 0.1M $MgSO_4 \cdot 7H_2O$.

2.1.2 Instruments/Equipment

The materials that were used for the research work are Canberra Model 727/727R Lead Shield Gamma-ray Spectrometer with NaI(Tl) detector, Oven Gallenham England, Hanna pH meter 209 Romania, Labtect digital conductivity meter USA, Beakers, Mortar and pestle, volumetric flask,

2.2 Area of Study

Barkin Ladi and Jos South Local Government Area of Plateau State are situated in the central part of Northern Nigeria, on a rugged terrain of lowlands. Five samples were collected each from Barkin Ladi and Jos South, Local Government Area. These areas are the principal centres of tin and columbite mineralization, which forms the focal area of younger granites. The mine tailings are associated with radioactive minerals as impurities such as monazite and zircon among others. These minerals are radioactive and can cause serious hazards to human health during mining and land cultivation for crop production.

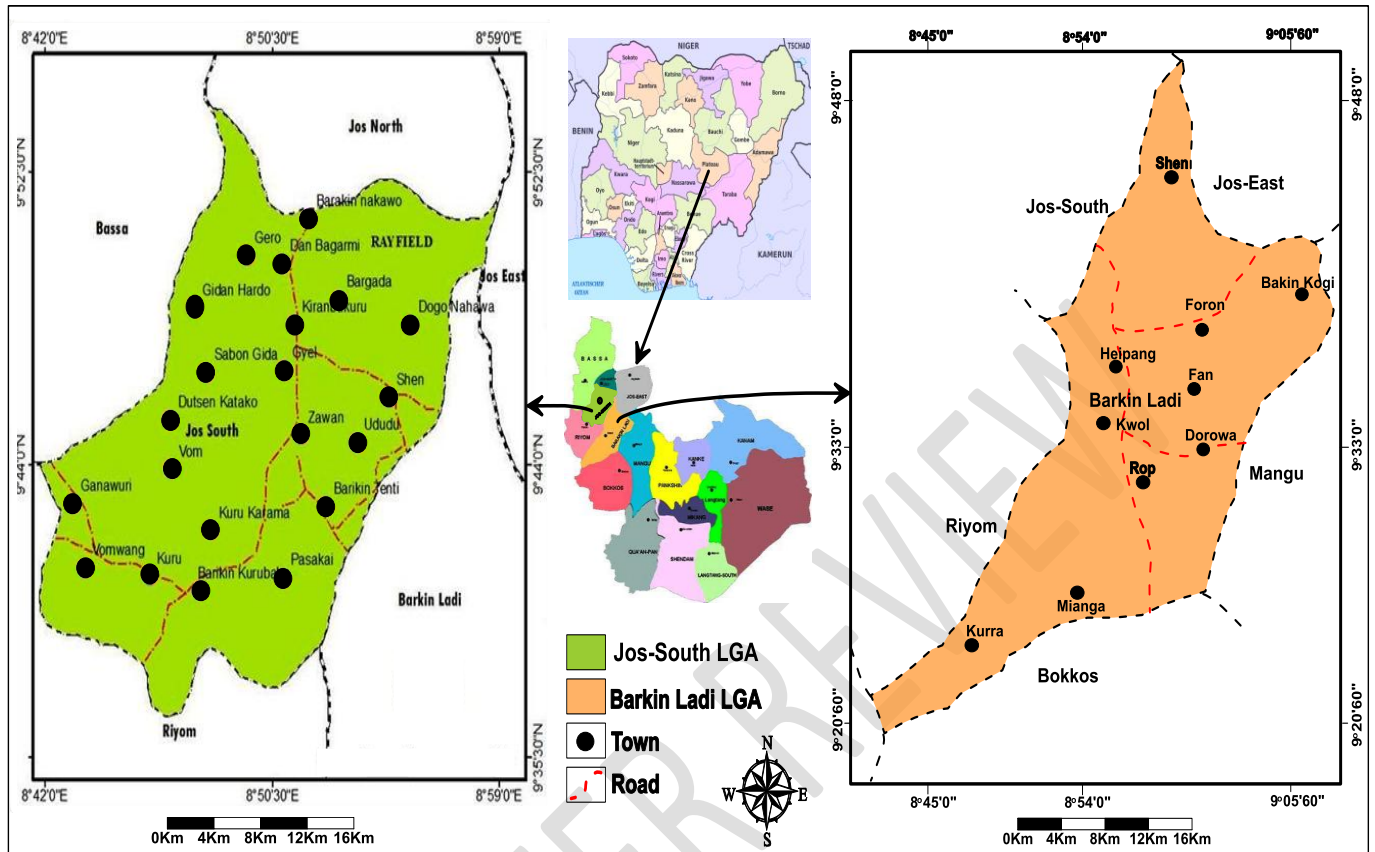


Figure: 1 Map of Barkin Ladi and Jos South Local Government Area

2.3 Sample Collection

2.3.1 Soil Sample

At each sampling point, about 0.50 kg of the soil sample was collected from a depth of between 0- 20 cm from the surface of the soil, using a clean auger at a distance of 1m away from each other, and within an area of one square meter.

2.4 Sample Analysis. A gamma ray Spectrometer with NaI(Tl) detector, was used to analyze the soil samples for the presence of ^{226}Ra , ^{232}Th , and ^{40}K . Gamma-ray (γ -ray) spectroscopy is a quick and nondestructive analytical technique that can be used to identify various radioactive isotopes in a sample. In gamma-ray spectroscopy, the energy of incident gamma rays is measured by a detector. By comparing the measured energy to the known energy of gamma rays produced by radioisotopes, the identity of the emitter can be determined. This technique has many applications, particularly in situations where rapid nondestructive analysis is required.

RESULT AND DISCUSSION

3.1 Results

Table 1: Summary of the result for the analysis of radionuclides ^{40}K , ^{226}Ra , and ^{232}Th sample collected in Barkin Ladi and Jos South LGA

SAMP LE ID	^{40}K	^{226}Ra	^{232}Th	pH	OC %	OM %	CEC cmol /kg	Clay %	Silt %	Sand %
KW ₁	210.26±0.24	98.71±0.64	93.79±0.26	4.10	0.32	0.07	4.36	30.72	22.00	47.28
KW ₂	276.08±0.08	97.32±0.40	39.08±2.40	4.11	0.24	0.09	4.52	40.12	12.42	47.46
KW ₃	124.97±0.31	51.99±0.30	26.99±0.38	3.21	0.42	0.38	2.61	30.53	25.76	43.71
FJ ₁	315.23±0.25	98.21±0.41	89.24±0.33	3.76	0.28	0.02	4.49	36.72	24.00	39.28
FJ ₂	204.14±0.32	85.88±0.24	72.48±0.22	3.93	0.21	0.21	3.21	42.96	23.91	33.13
FJ ₃	142.87±0.46	67.87±0.12	64.23±0.10	6.81	0.23	0.01	4.91	29.32	38.21	32.47
B ₁	231.86±0.34	85.97±0.41	69.08±0.51	4.34	0.20	0.41	2.03	30.72	28.00	41.28
B ₂	249.41±0.24	63.43±0.52	62.21±0.55	3.76	0.20	0.32	3.50	25.21	22.56	52.23
B ₃	210.27±0.10	46.19±0.80	41.88±0.08	5.32	0.30	0.62	2.90	21.24	40.39	38.37
NK ₁	301.91±0.24	83.41±0.30	76.20±0.37	5.16	0.16	2.97	1.29	20.72	24.00	55.28
NK ₂	236.31±0.33	58.02±0.35	58.57±0.43	5.10	0.24	3.01	3.32	40.78	23.67	34.54
NK ₃	139.92±0.24	43.10±0.62	35.73±0.51	4.21	0.41	1.41	2.90	10.02	35.32	54.66
N ₁	208.11±0.34	105.42±0.1	86.65±1.73	3.96	0.16	2.96	3.61	34.61	22.41	42.98
N ₂	157.12±0.48	99.91±1.11	67.39±0.94	4.50	0.08	3.12	1.01	20.50	38.67	38.83
N ₃	167.32±0.15	57.02±0.22	61.20±0.22	6.32	0.19	2.74	4.90	36.00	12.19	51.81
K ₁	259.34±0.44	62.54±0.36	31.94±0.51	4.61	0.24	0.45	1.16	9.72	13.00	76.28
K ₂	172.15±0.12	53.32±0.18	47.61±0.41	5.28	0.04	1.84	3.76	31.87	19.38	48.75
K ₃	161.39±0.18	44.32±0.32	36.92±0.54	6.71	0.31	2.59	2.41	30.97	22.43	56.60
SSK ₁	305.49±0.31	79.13±0.81	98.40±0.52	4.06	0.40	0.10	1.34	10.72	14.00	75.38
SSK ₂	201.64±0.37	62.51±0.37	47.38±0.42	4.32	0.24	1.93	3.21	29.67	36.41	33.92
SSK ₃	209.74±0.22	35.31±0.33	37.18±0.57	3.70	0.50	2.03	3.82	31.90	21.87	46.23
Z ₁	319.48±0.22	69.65±0.13	52.46±0.23	4.34	0.32	1.05	2.09	35.87	22.65	41.48
Z ₂	211.94±0.14	43.78±0.14	23.87±0.40	3.12	0.12	3.32	3.52	25.12	26.57	48.31
Z ₃	167.92±0.32	32.47±0.21	16.99±0.23	4.21	0.02	2.04	4.01	23.43	29.31	47.26
ZPSC ₁	261.14±0.39	77.32±0.41	27.23±0.49	6.90	0.39	2.01	4.11	38.00	12.45	49.55
ZPSC ₂	200.27±0.82	52.34±0.82	22.43±0.73	4.92	0.12	1.52	1.08	20.35	40.12	39.34
ZPSC ₃	159.38±0.07	31.39±0.73	24.35±0.20	5.80	0.31	2.04	2.90	25.56	40.32	34.12
KJ ₁	180.39±0.42	109.61±0.0	102.50±0.5	3.82	0.40	1.10	1.27	10.72	14.00	74.28
KJ ₂	263.78±0.67	77.27±0.44	66.72±1.92	3.56	0.44	2.01	4.10	40.82	8.32	50.86
KJ ₃	210.98±0.25	31.67±0.68	42.65±0.00	5.63	0.51	0.63	2.32	30.10	15.30	54.60
Permissible limits	420	33	45							

N=3

Key: kw=kwat, FJ= Foron junction, B=Ban, Nk=Nkan, N=Newo, K=Kuru, SSK=Science School Kuru, Z= Zawan, ZPSC=Zawan Police Staff College and KJ=Kuru Jenta

OC = Organic Carbon, OM = Organic Matter, CEC= Cation Exchange Capacity

NB: 1 stands for samples collected directly from the mining spot, 2 stands for samples collected 200 m away from the mining spots, and 3 stands for samples collected 400 m away from the mining spots

The activity concentration of ^{40}K , ^{226}Ra , and ^{232}Th expressed in Bq/Kg for the samples obtained from mining areas of Foron and Heipang district of Barkin Ladi Local Government Area and

Kuru and Zawan district of Jos South Local Government area of Plateau State are presented in Table 1 above. The samples were taken during the second week of July 2021 at the various sites listed in Table 1 above. A total of thirty (30) samples were taken from various sampling points. From each of the settlements, three samples were taken. The samples were taken from the point source, 200 meters and 400 meters away, respectively. This was carried out to evaluate the distribution and fate of radionuclides from the environment in the mining locations. The radioactive content in the samples from the various sites varied significantly. The findings show that the concentration of radionuclides near mining sites was higher than that at 200 meters away from the sites and at 400 meters away respectively. With a few exceptions where the opposite was true, these patterns were seen almost everywhere during the sampling. At a 200-meter distance from the mining sites, KW₂ had a greater concentration of ⁴⁰K than KW₁ (i.e. point source). B₂ samples, which were taken 200 meters away from the point source, were likewise more abundant than B₁ samples collected from mining locations. SSK₃ was greater than SSK₂, as well. Given that ⁴⁰K is a nutrient for plants, the cause of this fluctuation may be related to the physicochemical characteristics of the soil and the surrounding vegetation. The overall results for ²²⁶Ra and ²³²Th exhibit a pattern, with the higher values (results) coming from samples taken directly from the mining sites, followed by samples taken 200 and 400 meters distant from the sites, respectively. These findings show that radionuclides are typically released into the environment through a variety of activities, some of which may be connected to mining processing, transportation, water erosion, and wind. A significant additional factor that influenced the distribution and fate of radionuclides in the soil is the properties of the particular radionuclide, its chemical form and reactivity, control of the nature of its retention in the soil, and the affinity to certain soil constituents [10]. The concentration of ²²⁶Ra varies from 109.61 ± 0.04 to 62.54 ± 0.36 in mining sites, 99.91±1.11 to 43.78±0.14Bq/kg from 200m away from mining sites, 67.87±0.12 to 31.39±0.73Bq/kg from 400m away from mining sites respectively. These values are similar to those reported by [11], in Mazat Barkin Ladi. Almost all the results recorded are higher than the permissible limits of 33Bq/kg except for a few cases in KJ₃, ZPSC₃, and Z₃ which fall below the recommended limits of 33Bq/kg [12]. ²³²Th ranges from 102.50±0.55 to 27.23±0.49 around the mining sites, 72.48±0.22 to 22.43±0.73 200m away from the mining sites and 64.23±0.10 to 6.99±0.23 Bq/kg 400

meters away respectively. K_2 and K_3 recorded a higher value than K_1 . Soil physicochemical properties may also have been responsible for this abnormality. All the results of ^{232}Th around the mining sites are higher than 45 Bq/kg permissible limits, except for K_1 and ZPSCs₁ whose values are 31.94 ± 0.51 and 27.23 ± 0.49 .

The most significant property of soil is its pH level, it has effects on all other parameters of soil. Therefore, pH is considered while analyzing any kind of soil. If the pH is less than 6 then it is said to be an acidic soil. When pH ranges from 6 to 8.5, it is said to be normal soil, but greater than 8.5 then it is said to be alkaline soil. [13]. The pH of the soil samples from this investigation ranges from 3.12. to 6.8 which is lower than the 7.3 to 7.9 for raining season reported by [14], but similar to the report of [15]. A decrease in the pH values will lead to an increase in the mobility of the radionuclides [16]. Additionally, in this study, the organic carbon (OC) ranges from 0.51% to 0.02%, and Organic Matter ranges from 3.33% to 0.01%. Soil organic matter (OM) is of great importance due to its influences on the mobility, solubility, and complexation of the radionuclides in the soils [15]. Cation exchange capacity ranges from 4.91Cmol/Kg to 1.01Cmol/kg respectively. Other soil properties studied were clay content which ranges from 42.96 % to 10.02%, which is similar to the report of [15]. A decrease in clay content will increase the chances of the mobility of the radionuclides. Silt ranges from 40.39 % to 8.32% and sand from 76.28 to 32.47% higher than the report of [15]. A decrease in sand content will lead to a decrease in the mobility of the radionuclides and vice versa [16].

Generally, the high concentration of radionuclides may be a result of the application of phosphate fertilizer in the farmland around the study area[17].

Table 2: Mutual correlation matrix of radionuclides and soil properties of Jos South and Barkin Ladi Local Government Area.

	^{40}K	^{226}Ra	^{232}Th	pH	OC	OM	CEC	Clay	Silt	Sand
^{40}K	1									
^{226}Ra	0.854	1								
^{232}Th	-0.510	0.701	1							
Ph	0.633	0.918	0.506	1						
OC	-0.910	-0.901	-0.350	-0.840	1					
OM	-0.894	-0.986	-0.767	-0.840	0.870	1				
CEC	0.884	0.996	0.650	0.911	-0.933	-0.982	1			
Clay	0.935	0.887	0.348	0.800	-0.997	-0.867	0.923	1		
Silt	-0.444	-0.514	0.236	-0.701	0.756	0.393	-0.559	-0.726	1	
Sand	-0.912	-0.771	-0.743	-0.457	0.687	0.864	-0.778	-0.722	0.049	1

To draw the provenance of radionuclides and their relationship with the soil physiochemical properties, a Pearson correlation matrix was tabulated in Table 2 above. ^{40}K shows a strong positive correlation with ^{226}Ra ($r = 0.854$, $p \leq 0.05$), which signifies their common mineralogical affinity and/or similar sources, it also signifies that the absence of one will affect the other. Both ^{226}Ra and ^{40}K are released from parent minerals (e.g. clay minerals) as ions during weathering, and thereafter preferentially adsorbed by the clay [18], from the clay, Ra and K are transferred to the soil solution and are available for migration and uptake by plants/vegetables [19]. A moderate negative correlation was observed between ^{40}K and ^{232}Th ($r = -0.510$, $p \leq 0.05$), which indicates their different geochemical behaviour [18][19]. This negative correlation was similar to the report of Habib *et al.* (2018). ^{232}Th shows a very strong positive correlation with ^{226}Ra ($r = 0.701$, $p \leq 0.05$). pH of the soil shows a strong positive correlation with ^{226}Ra ($r = 0.918$, $p \leq 0.05$) which is an indication that the solubility and mobility of ^{226}Ra increase with increasing soil acidity [20]. ^{40}K was negatively correlated with OC and OM ($r = -0.910$, $p \leq 0.05$ and -0.894 , $p \leq 0.05$), which reveals inorganic mineralogical (e.g. illite) affiliated with OM [19]. Clay has a strong correlation with ^{40}K and ^{226}Ra ($r = 0.935$ and 0.887), while a weak correlation was observed between Clay and ^{232}Th ($r = 0.348$). CEC has a strong positive correlation with Clay ($r = 0.923$). The reason may be a result of the fact that clay minerals exhibit the largest surface area, which is important for soil chemistry and CEC, and also for water-holding capacity important for transporting nutrients and pollutants to soil organisms and plants [16]. Sand witnessed a strong negative correlation with CEC ($r = -0.722$), this is because of the inability of sand to retain some nutrients and some inorganic elements.

One-way analysis of variance (ANOVA) was performed to compare the radionuclide concentration and their fate and migratory patterns in the environment based on the different soil physiochemical properties. The one-way ANOVA revealed that there was a statistically significant difference in the mean concentration of the radionuclides (F ratio values was 218.9890) $p = 0.00001$ at $p = 0.05$

4.0 Conclusion

Natural radionuclide distribution into the environment and statistical analysis of the naturally occurring radioactive materials in soils of some abundant mining sites of Jos South and Barkin Ladi

Local Government Area of Plateau State Nigeria were studied. It was found that the natural radionuclides of ^{226}Ra , ^{232}Th , and ^{40}K are the main radiological constituents of the soil. The result of the analyses shows that the activity concentration of the natural radionuclide in some of the sampling sites is above the recommended limits whereas some are lower. The result for ^{40}K in all the soil samples is below the permissible limits. The result reveals that the activities of artisanal miners around the ex-mining site contribute to the higher concentrations of radionuclides in most of the areas, and also aid in the distribution of the radionuclides into the environment.

Reference

1. Heikal, M. T. S., Monsef, M. A. E., Goma, S. R., Mansi, M. M. E., & Top, G. (2018). Natural Radionuclides Levels and their Geochemical Characteristics of Abu Dabbab Albite Granite Mining Area, Central Nubian Shield of Egypt. *Journal of Environmental Hazards* 1(1). 1-14
2. Banari, B.S. (2015). The Calculation of the Annual Effective Dose Due to Exhalation of Radon Progeny in Iran. *Journal of Applied Environmental and Biological Sciences*, 5: 609–613
3. Obaid, S.S., Gaikwad, D.K. and Pawar, P.P. (2015). Determination of natural radioactivity and hazard in some rock samples. *Bionano Frontier*, 8(3), 125–127
4. United Nations Scientific Committee on the Effects of Atomic Radiation and Annex, B. (2000). Exposures from natural radiation sources. Cosmic rays, 9 .11.
5. Najam, L.A., Younis, S.A. and Kithah, F.H. (2015). Natural radioactivity in soil samples in Nineveh Province and the associated radiation hazards. *International Journal of Physics*, 3(3): 126–132.
6. Taqi, A.H., Shaker, A.M. and Battawy, A.A. (2018). Natural radioactivity assessment in soil samples from Kirkuk city of Iraq using HPGe detector. *International Journal of Radiation Research*, 16(4), 455–463.
7. Thabayneh, K. (2012). Natural radioactivity levels and estimation of radiation exposure in environmental soil samples from Tulkarem Province–Palestine.
8. Baba, A., Bassari, A., Erees, F. and Cam, S. (2004). Natural radioactivity and metal concentrations in soil samples taken along the Izmir-Ankara E-023 highway, Turkey.
9. Cengiz, G.B. and Öztanriöver, E. (2018). Analysis of natural radioactivity levels in soil samples and dose assessment for Digor District, Kars, Turkey. *Caucasian Journal of Science*, 5(1):30–39.
10. Seaman, J.C & Kimberly, A.R (2012). Radionuclide Fate radionuclide fate and Transport in Terrestrial Environments radionuclide transport in terrestrial environments. *Encyclopedia of Sustainability Science and Technology*; pp.8597-8634
11. Ogori B.O., Lubis S and Bakji G (2023). Determination of the Concentrations of Radionuclides and Heavy Metals and Their Transfer Factor from Soil to Crops/Vegetables in Some Agricultural Soils in Barkin Ladi Area, Plateau State, Nigeria. *International journal of research and innovation in applied science* 8 (5): 124-130
12. UNSCEAR. (2008): Sources and Effects of Ionizing Radiation: Report to the General Assembly, With Scientific Annexes, 2, 1–219. United Nations, New York;
13. Kekane, S.S., Chavan, R.P., Shinde, D.N., CL Patil, C.L. and Sagar, S.S (2015). A review on physicochemical properties of soil. *International Journal of Chemical Studies* 3(4): 29-32

14. Olatunji O, A., Komolafe E. T., Oke, S.O. (2016). Seasonal Variation in Physicochemical Properties of Soil within the Vicinity of an Iron Smelting Factory - Implication on Standing Vegetation. *Notudea Scientia Biologicae*, **8**(2):220-225.
15. Habib, M.A., Basuki, T., Miyashita, S., Bekelesi, W., Nakashima, S., Phoungthong, K., Khan, R., Rashid, M.B., Md, A.R., Isalm, T and Techato K. (2018). Distribution of naturally occurring radionuclides in soil around a coal -based power plant and their potential radiological risk assessment. *Radiochimica Acta*. **2**: 1-16
16. Smiciklas, I and Sljivic-Ivanovic M. (2016). Radioactive Contamination of the Soil: Assessments of Pollutants Mobility with Implication to Remediation Strategies. **Soil Contamination**. IntechOpen. Rijeka: chapter 13
17. Adjirackor, T., Darko, E. O and Sam F.(2017). Naturally occurring radionuclide transfer from soil to vegetables in some farmlands in Ghana and statistical analysis. *Radiation Protection Environment*, **40**: 34-43
18. Foth, H. D. (1984). Soils and minerals nutrition of plants, Fundamentals of soil science, 7th ed. Wiley. New York
19. Kabata-Pendias , A. and Henryk, P.(2001). **Trace element in soils and plants** 3rd ed. BocaCRC Press, Raton, Fla, London
20. Papaefthymiou, H.V., Manousakas, M., Fouskas, A. and Siavalas, G. (2013). Spatial and vertical distribution and risk assessment of natural radionuclides in soils surrounding the lignite-fired power plants in Megalopolis basin, Greece, *Radiation. Protection. Dosimetry***156**(1) 49