Investigation of the Transfer Factor and Dose Rate of radionuclides in Some Selected Crops Within Nasarawa State Nigeria.

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ABSTRACT

The activity concentrations of natural radionuclides, dose rate and soil-to-plant transfer factor have been evaluated in soils and crop samples from twenty locations in Nasarawa state Nigeria with the aid of a P- type, High Purity Germanium (HPGe) detector. The mean specific activities of ^{40}K, ^{232}Th and ^{238}U in the soil samples were 408.69, 24.08 and 30.71 Bq kg^{-1}, respectively, while the average activity concentration of ^{40}K, ^{232}Th and ^{238}U in crop samples were 142.63 Bqkg^{-1}, 46.06 Bqkg^{-1} and 17.45 Bqkg^{-1}, respectively. The radium equivalent concentration, the external and internal hazard indices were estimated, and ranged from 81.77 to 159.09 Bqkg^{-1}, 0.22 to 0.43, and 0.28 to 0.53, with average values of 115.50 Bqkg^{-1}, 0.31 and 0.40, respectively. The average soil-to-plant transfer factors for ^{40}K, ^{232Th} and ^{238}U were 0.053, 0.369 and 0.366, respectively. The mean absorbed dose and the mean annual outdoor effective dose equivalent in soil samples were 105.88 nGyh^{-1} and 0.13 mSvy^{-1} respectively. The mean annual effective dose equivalent for the study area is higher than the world average (0.07mSvy^{-1}) and international recommended standards of 0.1 mSvy^{-1} recommended by World Health Organization (WHO). The excess lifetime cancer risk (ECLR) ranged from 0.17 $\times10^{-3}$ to 1.16 $\times10^{-3}$, with a mean of 0.46 $\times10^{-3}$. This value is higher than the world average of 2.9 $\times10^{-4}$ reported by United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR. This value could pose a seriouse radiological hazard both the farmers and the general public. ^{238}U recorded the highest soil-to-plant transfer factor compared to other radionuclides in the soil, this may be due to its higher accumulation in soil and higher uptake by crops. Regression analysis showed that there was no linearity in the relationship between activity concentration of radionuclides in soil and in crops.

1.0 Introduction

Natural radionuclides are present in every human environment such as soil, rock, plants, water, air, foods and even the human body contains naturally occurring radioactive materials (NORM). The radionuclides present in the environment are normally in very low activity concentrations. The sources of these radionuclides are natural as well as man-made. The natural radioactive background originates from uranium and thorium series, from potassium - 40 and from the interaction of cosmic radiation with matter, while man-made sources include various applications of radionuclides in medicine, industries, agriculture, consumer products and nuclear weapon tests (UNCEAR, 2000).

Food crops from contaminated environment may accumulate radioactivity that could form a direct route of exposure to human population when consumed. Therefore, the knowledge of radioactivity levels in grown food crops is very important in order to establish the dose received by populations. The main radioactive environmental contamination in Nigeria are from the natural radioactive materials that could enter the environment either through uncontrolled mining activities or from the use of phosphate fertilizers which are manufactured from phosphate rocks (Banzi et al. 2002).

The radioactivity level in soil can likely be used to show the magnitude of contamination in locally grown food crops, but it cannot describe the biological effects of radiation exposure to individuals who consume that food. Therefore the estimation of doses is usually carried out for assessing health safety of an individual undergoing radiation exposure through ingestion of contaminated food. The intake of radionuclide within food is dependent on the concentration of radionuclides in various food crops and on the food consumption rates. The risks associated with an intake of radionuclides in the body are proportional to the total dose delivered by radionuclides while staying in various organs. In general, it is assumed that stochastic effects occur linearly with dose and usually the annual effective dose quantities (AED) are usually used to define those risks when prolonged exposure to a single individual from a single intake of a radionuclide is being considered. Nasarawa state being an agricultural state with increasing

human activities and natural disaster, may have environmental pollution challenge due to the high level of mining activities, use of agrochemical, fertilizer application and indiscrimate waste disposal.

2.0 Determination of activity concentrations of radionuclides

The activity concentrations of ^{238}U, ^{232} Th, and ^{40} K in the samples were measured using p-type High pure Germanium (HPGe) gamma ray spectrometer: model GCM-8023; seriel number 9744, end cap diameter 78 mm, length 69.8 mm with efficiency of 80% and energy resolution of 2.3 KeV-FWHM at 1332 KeV peak of ^{60}Co.

2.1 Estimation of soil-to-plant transfer factor (TF)

A standardized soil layer was adopted in this work. The soil depth value was 20-25cm. Using IAEA guidelines, the soil-to-plant transfer factor *TF* was estimated as using (IURE, 1994) :

$$\mathrm{TF_p} = \frac{\text{Activity concentration of radionuclides in plant dry matter}}{\text{Activity concentration of radionuclides deposit in soil dry mass}} \quad (1)$$

where TF_p is the transfer factor of activity to plant .

The soil-to-plant transfer factors have been calculated for soil samples analysed for radioactivity to evaluate the rate of migration of radionuclides from soil to the plants in the study area.

2.3 Radium Equivalent activity (Ra_{eq})

The radium equivalent activity is an index that was introduced to represent the specific activity of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity which takes into account the hazards of radiation associated with the radionuclide elements. The expression used in estimating Ra_{eq} is given as (Araromi *et al*., 2016);

$$Ra_{eq} = 0.077A_k + A_U + 1.43A_{Th} \quad (2)$$

Where, A_k, A_u and A_{Th} are the average activity concentrations of ^{40}K, ^{226}Ra (^{238}U) and ^{232}Th respectively. From the above expression, it has been assumed that 4810 BqKg^{-1} is for ^{40}K, 259 Bqkg^{-1} for ^{232}Th and 370 Bqkg^{-1} for ^{226}Ra. ^{238}U and ^{226}Ra produce the same gamma dose; hence the assumed activity value is used for ^{226}Ra and ^{238}U interchangeably.

2.4 Radiation hazard index

The internal and external hazard indice is defined (Alharbi and El-Taher , 2013) as;

Internal hazard index; $$H_{int} = \frac{A_k}{4810} + \frac{A_u}{185} + \frac{A_{Th}}{259} \leq 1 \quad (3a)$$

External hazard index; $$H_{ext.} = \frac{A_k}{4810} + \frac{A_u}{185} + \frac{A_{Th}}{259} \leq 1 \quad (3b)$$

The external hazard index is obtained from the expression of Radium equivalent activity via the assumption that its maximum allowed values corresponds to the upper limit of Ra_{eq} (370 $Bqkg^{-1}$) in order to limit the radiation dose of 1.5 msv/y. if the radiation hazard must be insignificant, then the index value must be less than one. Once the value is above one, the area needs a proper check to save the lives of the people living or working with the area.

2.5 Dose evaluation

Exposure and absorption of radiation at 1 m above the ground containing naturally occurring radionuclide poses radio- logical hazard to human. The total absorbed dose rate in air, (ADRA or D in nGy h^{-1}) was calculated using (UNSCEAR, 2000);

$$D\ (nGyh^{-1}) = 0.462A_u + 0.604A_{Th} + 0.0417A_k \quad (4)$$

where A_K , A_u and A_{Th} are the specific activities (in Bq kg^{-1}) for ^{40}K, ^{238}U and ^{232}Th respectively in soil samples and the concentration-to-dose conversion factors are 0.0417, 0.462 and 0.604, respectively.

2.6 Annual effective dose rate (AEDR)

The annual effective dose rate of any considered population was estimated using the conversion coefficient from the absorbed dose in the air to the effective dose given as (0.7SvG y^{-1}) and taking into account the outdoor occupancy factor (0.2), and the indoor occupancy factor (0.8) (UNSCEAR, 1998). The outdoor AED is obtained from the expression (UNCEAR, 2000):

$$Outdoors\ AED\ (mSvy^{-1}) = AD(nGyh^{-1})X\ (8760h)\ X(\ 0.7SvG^{-1})X\ (0.2) \quad (5)$$

The total annual effective dose rate (AED) is the sum of the annual effective dose rate of natural radionuclides and the annual effective dose rate of radionuclides in (μSv/y).

The excess lifetime cancer risks, which is the probability of an individual developing cancer over a lifetime when considering a specified exposure level was also calculated. The value obtained is the representation of the number of cancers that could be expected from a given number of people when exposed to a carcinogen at a given dose. The excess lifetime cancer risk (ELCR) was estimated using (Taskin *et al.,* 2009);

$$LTCR = AEDR(\frac{\mu sv}{y}) * AL * RF \quad (6)$$

Where; LTCR is life time cancer risk, AEDR is the annual effective dose rate, AL average life time RF is the risk factor (0.05) and the world average of LTCR was pegged at 0.299 $x10^{-3}$ (ICRP, 1990).

3.0 Materials and methods

3.1 Study area

Nasarawa State is bounded in the north by Kaduna State, in the west by the Federal Capital Territory, in the south by Kogi and Benue States and in the east by Taraba and Plateau States. It has a central location in the middle belt region, it lies between 7º45' and 9º25'N of the equator and between 7º and 9º37'E of the Greenwich meridian. The state has a total land mass of 27,137.8 square kilometer and a population of about 2,523,395 according to 2011 population estimate (NPC, 2011). Nasarawa state has 13 local governments, detail of coordinates and map is as contained in table 1 and figure respectively.

Table 1. Sample locations and coordinates

S/N	Location	Latitude	Longitude	Population
1	Lafia	8º29'38.04"N	8º30'55.15"E	371,986
2	Keffi	8º50'47.44"N	7º52'24.74"E	104,350
3	N/Eggon	8º44'33.029"N	8º32'30.804"E	167,971
4	Doma	8º23'35.05"N	8º21'19.58"E	156,712

National population census (2011)

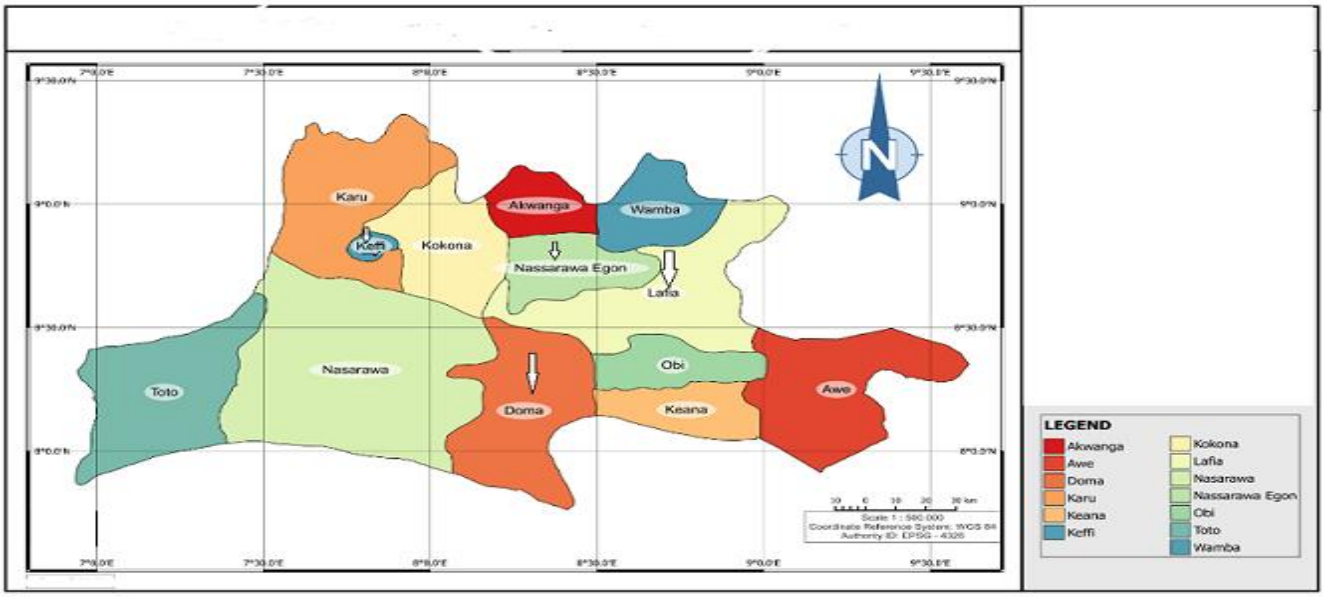

Figure 1: Map of Nasarawa state showing the local government areas of study.

3.2 Collection of samples

Using control tool, soil sample were collected to a depth of about 20-30 cm in accordance with the prescribed uniform practice (IAEA, 2010). This is to include the surface layer corresponding to the rooting zone. The soil sample was thoroughly mixed to provide a representative sample for each zone. A total of forty (40) samples were collected for both soil and crops

3.3 Sample preparation and measurement

A total of twenty (20) different soil samples were collected. The surface soil samples were collected (about 20-30 cm deep) from each location and about 1 kg of soil sample was packed in a polythene bag tied and labeled. The soil samples was pounded, crushed to a fine grain of about 100 meshes and sieved (removing large pieces) and mixed for homogenization. The Samples was then dried at about 110 ˚C for 24 hours to ensure that moisture is completely removed and until a constant weight is obtained. A mass of about 500 g of each soil sample from each sampling point were placed in a marinelli beaker, sealed and stored for a period of four weeks at room temperature to allow secular equilibrium between parent nuclides and daughter nuclides prior to gamma spectroscopy (Ghose *et al.*, 2012; Amrani and Tahtat, 2001; Khan and Khan, 2001; Kumar *et al.*, 2003)

Similarly, the crops were also collected. About one kilogram of crop was collected from each point and packed in polythene bags, labeled and taken to the processing room (laboratory). The crops were collected as they mature.

A total of twenty (20) food crops (yam, maize, beans and cassava) were collected and emptied into a polythene bag; tied and labeled. The food crop that are tubers were first washed under running water and then with distilled water to remove all the attached sand and dust particles and peeled for yam and cassava crops and then dried in air. The samples were further weighed and dried in the oven at about 110 ^{o}C for 24 hours to obtain constant dry weight. Samples were packed and grounded for homogenization.

About 500 g of each sample were packed into one liter of Marenilli beaker and sealed for four (4) weeks to reach secular equilibrium between parent nuclei with their daughter nuclei. This was done in order to allow for all radionuclides and its short-lived progenies to reach secular radioactive equilibrium prior to gamma spectroscopy.

Samples were placed in the HPGe detector for analysis to help identify the various nuclides present in each of the samples (soil and crops). With the aid of the detector and the use of AAS analyzer, the migration of radionuclides was evaluated by taking note of the type of heavy metal present and the duration plant has spent in the soil. From the detector too, the activity concentrations of both soils and crops were determined.

4.0 Results and discussion

The results of the average activity concentrations in both soil and crops were evaluated and the result is as contained in Tables 2 and 3 respectively. On the other hand, Tables 4 and 5 presents average transfer factor of radionuclides from soil to crops and radiological hazard indices of soil samples in Nasarawa state.

Table 2: Average activity concentration of soil sample in Nasarawa state

LGA	N	^{238}U(BqKg^{-1})	^{232}Th(BqKg^{-1})	^{40}K(BqKg^{-1})
Lafia	5	24.8007	16.75	309.395
Keffi	5	26.8233	22.115	569.33

N/eggon	5	22.91	27.82	389.7075
Doma	5	48.2967	29.6267	366.3375
Mean		30.7077	24.0780	408.6925

Table 3: Average activity concentration of crop sample in Nasarawa state

LGA	N	$^{238}U(BqKg^{-1})crp$	$^{232}Th(BqKg^{-1})crp$	$^{40}K(BqKg^{-1})crp$
Lafia	5	16.78	8.67	142.6375
Keffi	5	17.28	16.67	221.135
N/eggon	5	18.045	17.08	126.975
Doma	5	17.7	141.8	19.6
Mean		17.45125	46.055	127.586875

Table 4 : Average Transfer factor from soil to crops of sample area

LGA	TF(U)	TF(Th)	TF(K)
Lafia	0.73968	0.482202	0.329113
Keffi	0.644215	0.753787	0.388413
N/Eggon	0.453478	0.363288	0.335358
Doma	0.366485	0.369431	0.053503

Table 5:Radiological hazard indices of soil samples Nasarawa state

Locations	Absorbed dose(nGy/h)	AEDR(uSv/h)x10^{-3}	Ra_{eq}(Bq/kg)	H_{ext}	$H_{int.}$	LTCR (x 10^{-3})
Lafia	40.063501	0.049161922	81.7687	0.2208	0.2822	0.172066727
Keffi	50.344896	0.061778222	102.2862	0.2763	0.3487	0.216223777
N/Eggon	63.13782906	0.07747643	159.0914	0.4296	0.5261	0.271167505
Doma	269.9741547	0.331285285	118.8708	0.3211	0.4516	1.159498498
mean	105.8800952	0.129925465	115.504275	0.31195	0.40215	0.454739127

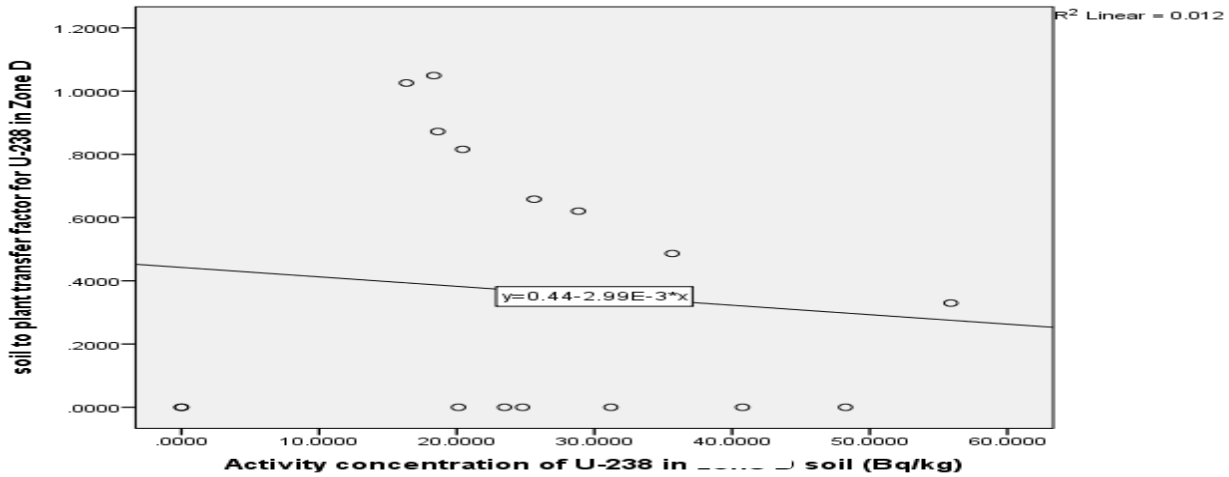

Figure 1a: Soil to plant transfer negatively correlated with activity concentration of ^{238}U in soil.

Figure 1b: Soil to plant transfer positively correlated with activity concentration of ^{232}Th in soil.

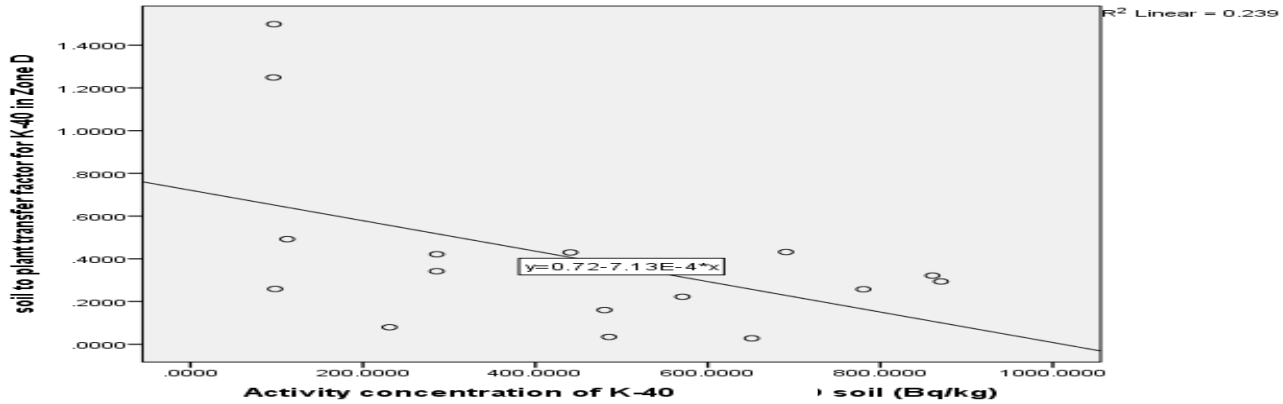

Figure 1c: Soil to plant transfer negatively correlated with activity concentration of ^{40}K in soil.

4.1 Activity concentration in soil and plant

The activity concentration of ^{40}K shows wide distribution from results of the study area. the activity concentration of ^{40}K ranging from 96.2 $BqKg^{-1}$ to 870.32 $BqKg^{-1}$ with an average value of 408.67 $BqKg^{-1}$, which a little above world average of 400 $BqKg^{-1}$, the values for ^{232}Th ranges from BDL to 550.67 $BqKg^{-1}$ with an average value of 24.08 $BqKg^{-1}$ which is below the world average and the activity concentration value for ^{238}U in the area ranges from BDL to 55.89 $BqKg^{-1}$. With an average value of 30.70 $BqKg^{-1}$, this is below the world average value.

4.2 Transfer factor

The average Transfer factor for the area, ^{232}Th leads in the range 0.36 to 0.75 and its closely followed by the value of ^{238}U having the range of value 0.36 to 0.76. ^{40}K has the lowest value of transfer factor in the zone with the range of value 0.05 to 0.38. The major factor that turns to affect the value of TF from the results obtained are soil, time and type of crop planted.

Linear uptake of elements by the plants was assumed in the estimation of transfer factors. This implies that the transfer factor for an element is constant under comparable conditions, irrespective of the element concentration in the soil (Tuovinen *et al.,* 2011) . Regression analyses were performed between activity concentrations of radionuclides in the soil and plant. The result did not show a linear relationship between the transfer factor and activity concentration in soil for the primordial radionuclides. There were cases where activity concentrations in plants were relatively constant or with narrower variation range at widely varying soil concentrations. This resulted in a large random variation between the two parameters. Ibikunle *et al.,* 2019, made a similar observation in their study on radiation dose and soil to plant transfer of radionuclides in south west Nigeria.

The same trend was obtained in this study. There is a non-linear relationship between the activity concentrations of radionuclides in the soil and plant. This may be because the uptake of an element by the plants can be influenced by various factors. It has been observed that soil moisture can affect the availability of some element in the soils. Also, the cation exchange

capacity, pH value, the organic matter content, nutrients in the soil and clay content, can seriously affect the availability of some element in the soils (Simon *et al.*, 2002). Considering the highlighted properties and their influence on radionuclide uptake and retention, the radionuclides that are present in higher concentrations are more likely to be retained in the soil, leading to a non-linear transfer to the plant. Notwithstanding, it may be possible to have a linear relationship between the activity concentration of radionuclides in soil and the corresponding transfer factor under a controlled farm and advance laboratory apparatus.

4.3 Radium equivalent activity

The result of the Radium equivalent activity of the area ranges from 81.77 $Bqkg^{-1}$ to 159.09 $Bqkg^{-1}$ with an average value of 115.50 $Bqkg^{-1}$ which is lower than the recommended maximum value of 370 $Bqkg^{-1}$ (UNSCEAR, 2000).

4.4 External and internal hazard indices (H_{ex} and H_{in})

The result obtained in all the study area, shows that the value for both H_{ex} and H_{in} are less than one and this is to show that the activity concentration of radionuclides present in the soil are within the recommended environmental safety limit. The approved value for H_{ex} and H_{in} must be less than unity (Otwoma *et al.*, 2013).

The gamma absorbed dose rates in the air for the study area ranges from 40.06 to 269.97 with an average value of 105.88, this is by far higher than the worldwide average value of 54 nGy h^{-1} (UNSEAR, 2008). The highest value was recorded in Doma local Government area of the state.

4.5 The annual outdoor effective dose equivalent

The result of outdoor annual effective dose equivalent is presented in the Table 6 has an average value of 129.93μSvy^{-1}. With the corresponding worldwide average value of 72.50 μSvy^{-1} (UNSCEAR, 2000], it can be observed that the mean annual outdoor effective dose equivalent for the study area is higher than world average and higher than 0.1 mSv y^{-1} maximum limit

recommended by WHO (WHO, 2006) but lower than 1mSvy $^{-1}$ limit recommended by NCRP (NCRP, 1975) and ICRP (ICRP, 1990). This indicates that there are higher risks of radiation induced cancer in some locations in the study area where there is higher effective dose than what was recommended for human radiological safety. This could be because of high agricultural activity and the use of soil enhancement chemical fertilizers and herbicides.

4.6 The excess lifetime cancer risk

It was observed that the samples having higher (ELCR) were collected from areas with high agricultural activities in Nasarawa state. Long-term exposure to radiation could have some risks of causing cancer. This implies that every individual has a risk of getting cancer at a time in his lifetime. The mean (ELCR) factor assessed in this work (0.46×10^{-3}) is higher than the world's average of 0.299×10^{-3} (UNSCEAR, 1988) , which implies that there is higher cancer risk in the area of study due to exposure to ionizing radiation from primordial radionuclides. Pakistan Institute of Medical Sciences (PIMS) Islamabad reported numerous cancer deaths cases from the Northern Pakistan. According to its report, the cancer death could be related to the higher radioactivity in the area (Qureshi *et al.*, 2014) . The value reported (0.46×10^{-3}) is very close to the result obtained in this work. This implies that cancer risk could be highly significant in the study area, especially in Doma local government area where the value is very high.

5.0 Conclusion

The activity concentration in soil and crops and the transfer factor of soil-to-plant has been evaluated for 20 locations in Nasarawa state, Nigeria to evaluate the radiological impact level across the area. The results show that there is high natural radioactivity level in soils is associated with high usage of inorganic fertilizers and herbicides for farming. The average value obtained for ^{40}K in the study area compares favourably with the allowed average of 400 Bq kg^{-1} (UNSCEAR, 2000) . There was no even distribution of radionuclides in crops across the study area. Activity Concentration in crop samples did not follow the trend that was obtained in soil samples. This is because activity concentration in crop samples does not depend solely on soil radioactivity; there are other channels of contamination of plants (UNSCEAR2000). The highest activity concentration in crop were observed in areas where there is likely to be over dependence of chemical-based fertilizer for agricultural purposes. Transfer factor is very high for ^{40}K most especially in areas where soil activity is low. This shows that transfer factor is not a function of

soil radioactivity. The soil-to-plant transfer factors were higher at a place of low soil concentrations. Non-linearity in the transfer of elements, both stable and radioactive, from soil-to-crops was significantly evident as was reported earlier by Tuovinen *et al* (2011*)*. There is a higher probability of lifetime cancer risk in the specific areas with high usage of inorganic fertilizers and herbicides in the study area.

References

Amrani, D. and Tahtat, M. (2001). Natural radioactivity in Algerian building materials. Applied Radiation Isot. 54: 687-689.

Alharbi, A. and El-Taher A. (2013). A study on transfer factors of radionuclides from soil to plant. *Life Science Journal* (2): 320-328.

Araromi O.I., Ojo A.O., Olaluwoye M.O. and Odefemi O.B. (2016). The concentration of Natural Radionuclides in soil samples from the practical year agricultural farmland, University of Ibadan. *IOSR Journal of applied Physics* 8: 60-68.

Ghose, S., Asaduzzaman, K.H, Zaman, N., (2012). Radiological importance of marble used for construction in Bangladesh. Radioprotection 47 (1), 105-118.

Khan, K. and Khan H.M., (2001). Natural Gamma emitting radionuclides in Pakinstani Portland cement. Applied Radiation Isot., 54,861-865.

International Commission on Radiological Protection (ICRP), Recommendations of the ICRP, Pergamon Press, Oxford, UK, 1990 ICRP Publication 60. Annals of ICRP 21 (1–3) .

IURE(International Union of Radio Ecologists). (1994). Handbook of parameter values for the prediction of radionuclide transfer in temperate environments. Technical reports series no. 364, IAEA, Vienna.

Kumar, A., Kumar, M. and Singh, S. (2003). Natural activities of 238U, 232Th and 40K in some Indian building materials. Radiation measurement (36): 465-469.

NCRP, Natural Background Radiation in the United States. Recommendation of the National Council of Radiation and Measurements, 1975 Report No. 45

Otwoma D., Patel J.P, Bartold S., Mustapha A.O, (2013) Estimation of annual effective dose and radiation hazard due to natural radionuclides in Mount Homa, Southwest Kenya, Radiat. Prot. Dosim 155 (4) (2013) 497–504 .

Qureshi A.A, Tariq S., Ud Din K., Manzoor K., Calligaris C., Waheed A. (2014), Evaluation of excessive lifetime cancer risk due to natural radioactivity in the rivers sediments of Northern Pakistan, J. Radiat. Res. Appl. Sci. 7 (8) 1–10 .

Simon S.L., Graham J.C, Terp S.D, (2002) Uptake of 40 K and 137 Cs in native plants of Marshall Islands, J. Environ. Radioactivity. (59) 223–243 .

Taskin H, Karavus M, Ay P, Topuzoglu A, Hidiroglu S, Karahan G (2009) Radionuclide concentrations in soil and life-time cancer risk due to gamma radioactivity in Kirklareli,Turkey. *Journal of Environmental Radioactivity*, ***100(1):*** *49-53.*

Tuovinen T.S , Roivainen P. , Makkonen S., Kolehmainen M., T. Holopainen , J. Juutilainen, (2011). Soil-to-plant transfer of elements is not linear: results for five elements relevant to radioactive waste in five boreal forest species, Sci. Total Environ. 191–197.

UNSCEAR (United nations scientific committee on the effects of atomic radiation) (2000). Exposures from natural radiation sources. Report to the General Assembly, with annexes, Annex-B, United Nations, New York.

UNSCEAR(United Nations Scientific Committee on the Effects of Atomic Radiation), (2008), Report to the general assembly, vol. 1, annex B.

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). (1998), Source, effects and risks of ionizing radiation, report to the General Assembly, with annexes, United Nations, New York, p. 95 .

WHO Expert Group , Burton Bennett, Michael Repacholi, in: Carr Zhanat (Ed.), (2006) Health Effects of the Chernobyl Accident and Special Health Care Programs: Report of the UN Chernobyl Forum Health Expert Group (PDF), World Health Organization, Geneva,p. 106. ISBN 978-92-4-159417-2 .