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Determination of Radionuclide Content in Agricultural Lands using ResRad Off-Site Computer Model in Nasarawa State, Nigeria

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Abstract:- Nigeria as one of the developing countries, has most of its farmlands contaminated by human activities like; mining, waste disposal, industrial waste, agricultural practices, use of inorganic fertilizers, sewage, sludge, and other anthropogenic activities. This research work evaluated the activity concentrations of natural radionuclides, and excess lifetime cancer risk (ELTCR) in soil and crop samples from twenty locations of farmlands in Nasarawa state, Nigeria. High purity Germanium (HpGe) detector was used to determine the activity concentration of radionuclides. The absorbed dose rate and ELTCR for 60 years were forecasted and evaluated using the ResRad off-site computer model. The mean activity concentrations 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, 46.06, and 17.45 Bqkg^{-1}, respectively. The Ra_{eq} concentration, the external, and internal hazard indices were evaluated 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, 0.31, and 0.40Bqkg^{-1}, respectively. The ELTCR ranged from 0.17 ×10^{-3} to 1.16 ×10^{-3}, with a mean of 0.46 ×10^{-3}. This value is above the average value approved by UNSCEAR 2000.Correlation analysis showed a strong positive correlation between activity concentration of radionuclides in soil and crop for ^{40}K and ^{232}Th and a weak correlation for ^{238}U due to soil type and microbial activities in most of the study area, the total cancer risks in the zone, particularly in Akwanga and Doma, were higher than the safety limits. Excessive use of organic fertilizer on farmland should be checked from time to time.

Keywords:- *Radionuclide, Res Rad Off-Site Model, Excess Lifetime Cancer Risk, Activity Concentration.*

I. INTRODUCTION

Terrestrial and cosmogenic radiations are the major sources of radioactivity in our world today, and it has affected the soil, crops, human, air, and water[1 – 2]. These radionuclides come from natural sources like^{40}K, thorium, and uranium series, other sources are due to the use of radionuclides by man in agriculture, mining, medical science, manufacturers, and the use of nuclear weapons [2 – 3]**.** Over time, the environment is beginning to suffer negative effects from these radionuclides due to an increase in several human activities to meet up with the daily increasing need of a man in various sectors.

The soil-to-plant transfer factor (TF) is very important when it has to do with the evaluation of the radionuclide activity concentration in crops, and that of an internal and external radiation dose as a result of consumption of food crops. Radionuclide TF of different radionuclides is influenced by some factors such as the physicochemical properties of the soil, the form in which the activity enters the soil, duration of radionuclide in soil, nature of soil, kind of the crop, agricultural practices, climate condition of the area, and the way the root tubers are prepared after harvested, like peeling root crops, washing, and exposure to sunlight [3 – 4].The level of radionuclide deposits within the soil can predict the degree of contamination in crops planted in a polluted area based on some physicochemical parameters analyses, but it cannot inform on the health effects of radiation exposure on individuals who consume these crops that have been polluted. Hence, the evaluation of doses is typically administered in investigating the health safety of persons undergoing radiation exposure through consumption of food that has been contaminated[4]. The absorption rate of radionuclide by a human via food consumption is a function of the number of activity concentrations in crops via root uptake, and the quantity of contaminated food consumed over time which eventually deposits these radionuclides into organs and body parts, resulting in health challenges.

Nasarawa State is an agricultural state with increasing human activities and natural disasters that may have environmental pollution challenges because of the high level of mining activities within the area, use of agro-chemical, fertilizer application for agricultural purposes, indiscriminate waste disposal, and erosion. Agricultural produce like yam, maize, cassava, etc., from polluted areas, may accumulate to become an immediate source of radiation to the community if not checked[5]**.** The State is rapidly growing in agriculture, industrialization, and human population[6]. Hence, the knowledge of radioactivity levels

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ingrown food crops within the study area is extremely important to determine the dose received by humans.

II. STUDY AREAS

Nasarawa State is located within the North central part of Nigeria. It is found on lat. 7° 45' and 9° 25' N of the equator, and long.7°, and 9° 37' E of the GM. The state is surrounded by many States; Kaduna State, Plateau State, Taraba State, Federal Capital Territory, Benue State, and Kogi State. The State features a total acreage of 26,875.59 Km^2 and the population of people in the area according to the population census report 2006, was about 1,826,883 having a density of67persons/km^2. The topography of the State is majorly hilly and plain lowlands. The climate condition of the area is typical of the Torrid Zone due to its location. The State has a high and low temperature of 81.7° F and 16.7° F respectively. The rainfall of the State ranges from 131.73 to 145cm, depending on the area.

Fig 1 Map of Nasarawa State Showing the Study Area

➢ *Collection of Samples*

Soil samples were collected to a depth of about 20-30 centimeters following the generally accepted method by IAEA, 2010. This is often to incorporate the surface layer like the rooting zones. The soil sample was well mixed into a uniform sample for the entire zone. A total of forty (40) samples were collected for both soil and crops.

➢ *Sample Preparation And Measurement*

- *Soil Samples*

A total of twenty (20) different soil samples were collected. The topsoil samples were collected (about 20-30 cm deep) from each location. About one kilogram of soil sample was packed and put in a nylon bag tied and labeled with masking tape. The soil samples were pounded to fine particles, sieved (removing large pieces), and mixed for homogenization. The Samples were dried at about 110˚C for twenty-four hours to get a steady weight. A mass of about 0.5 kg of the soil sample from each sampling point was placed using a Marinelli beaker, sealed, and stored for a period of twenty-eight days at 37°C to permit secular equilibrium between parent nuclides, and daughter nuclides before using the HPGe detector [7].

- *Crop Samples*

About one kilogram of every crop was collected from every location and put in nylon bags, labeled, and taken to the laboratory. These crops were collected as they mature. A total of twenty (20) food crops (yam, maize, beans, and cassava) were collected and emptied into a nylon bag, tied, and labeled. The food crop that isa tuber was first washed under running water to get rid of all the attached sand and mud particles before it was peeled and then dried in air. The Samples were further weighed and dried in the oven at about 110 °C for twenty-four hours to get constant dry weight[8]. Samples were packed and grounded for homogenization. About 500 g of every sample were packed into one liter of Marinelli beaker and sealed for four (4) weeks to succeed in secular equilibrium between parent nuclei with their daughter nuclei. This was wiped out to permit all radionuclides and their short-lived progenies to succeed in secular radioactive equilibrium before gamma spectroscopy. Samples were placed within the HPGe detector for analysis to identify the varied energy levels within the spectrum. With the help of the detector, the activity concentrations of radionuclides were evaluated.

➢ *Radiation Hazard Index*

The internal and external hazard indices are defined [9] as;

Internal hazard index(H_{int});

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

External hazard index ($H_{ext.}$);

$$H_{ext.} = \frac{A_k}{4810} + \frac{A_u}{370} + \frac{A_{Th}}{259} \leq 1 \quad (1b)$$

The$H_{ext.}$ index is obtained from the expression of Ra_{eq}via the idea that its highest allowed values agree with the upper limit of Ra_{eq} (370 $Bqkg^{-1}$) to limit the radiation dose of 1.5 mSv/y. The radiation hazard is considered significant only when the index value is up to one.

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➢ *Dose Evaluation*

- *Absorbed Dose Rate (ADR)*

The ADR in the air of farmlands was evaluated, considering the effect of radiation on humans above one meter from the ground of the polluted area. The expression used was given by [10]

$$AD\ (nGyh^{-1}) = 0.462A_u + 0.604A_{Th} + 0.0417A_k \quad (2)$$

Where A_K, A_u, and A_{Th} are the specific activities ($Bqkg^{-1}$) for ^{40}K, ^{238}U, and ^{232}Th, respectively. The conversion factors of concentration to dose are 0.0417, 0.462, and 0.604, respectively.

- *Annual Effective Dose Rate (AEDR)*

The yearly effective dose rate of any considered persons was estimated using the conversion coefficient from the absorbed dose rate in the air to the effective dose given as ($0.7uSvy^{-1}$) and taking into consideration the outdoor occupancy factor of (0.2), therefore the indoor occupancy factor (0.8) (11). The outdoor AED is obtained from the expression [11] ;

$$Outdoors\ AEDR\ (mSvy^{-1}) = AD(nGyh^{-1})X\ (8760h)\ X(\ 0.7SvG^{-1})X\ (0.2) \quad (3)$$

- *Excess Lifetime Cancer Risk (ELTCR)*

The ELTCR of a person living or working in a contaminated area is the probability of that person developing cancer over a lifetime considering the level of exposure to radionuclides. These determined using [12] ;

$$ELTCR = AEDR(\frac{\mu sv}{y}) * AL * RF \quad (4)$$

Where; ELTCR is the excess lifetime cancer risk, AEDR is the annual effective dose rate, AL is the average lifetime, RF is the risk factor (0.05). The world average of ELTCR was pegged at 0.299×10^{-3}.

➢ *Statistical Analysis*

Correlation analyses were also carried out between activity concentration in the soil and the activity concentration in food samples using SPSS software to determine the extent to which the variations in activity concentration in the soil affects the variation of activity concentration in food crops using the coefficient of determination. Regression analyses were evaluated to ascertain the linearity of activity of radionuclide in soil.

➢ *RESRAD OFF-SITE Computer Model*

To estimate the dose rate and cancer risk as a result of radionuclides from study areas, the RESRADOFF-SITE computer model was used. Some input parameters were considered; soil density, rainfall, irrigation rate, wind speed, aquifer level, precipitation rate, runoff coefficient, water table drop rate, and uncontaminated unsaturated zone thickness.

III. RESULTS AND DISCUSSION

➢ *Activity Concentration in Soil and Crop*

The average activity concentration of ^{40}K in soil shows wide distribution from the results of the study area. The activity concentration of ^{40}K ranges from 309.4 to 569.3 $BqKg^{-1}$, with an average value of 408.7 $BqKg^{-1}$, which is a little above the world average value of 400 $BqKg^{-1}$.^{232}Th ranges from 16.8 to 29.6 $BqKg^{-1}$,with an average value of 24.08 $BqKg^{-1}$, which is below the world average. The activity concentration for ^{238}U in the area ranges from 22.8 to 48.3 $BqKg^{-1}$, with an average value of 30.70 $BqKg^{-1}$(Table2), this is also below the world average value [13]. The lower value of radionuclide (^{232}Th, and ^{238}U) observed in this zone could be as a result of frequent flooding and erosion as observed and complained by the farmers of the area during interactions with them. The washing away of the topsoil could also reduce the level of radionuclides in the soil.

Activity concentration of radionuclides in crops for ^{40}K ranges from 16.56 to 298.22 $BqKg^{-1}$, with an average value of 127.59 $BqKg^{-1}$. The value for ^{232}Th ranges from 8.67 to 288.66 $BqKg^{-1}$, with an average value of 46.06 $BqKg^{-1}$, and the activity concentration of 238U ranges from 16.24 to 19.23 $BqKg^{-1}$, with an average value of 17.45 $BqKg^{-1}$(Table 3). The activity concentrations in crops were all above the safety limit for Crops in the tropical environment. This could be a result of the use of agrochemicals and fertilizers in farmland and possible contamination of crops from the environment (ambient radiation) and the type of soil [14], Doma area has more clay soils and that could contribute to uptake by Crops (Table 1). Comparing the output of the activity concentration in soil of the area to the activity concentration in crops with the approved limit by IAEA, the crops show higher values of radionuclide absorption. This implies that the levels of radioactivity in crop samples in the study areas may pose a radiological hazard when such food is ingested directly.

Table 1 Physcio-Chemical Properties of Agricultural Soils in Nasarawa State

Locations	Clay(%)	Silt(%)	Sand(%)	pH level	Organic matter (%)
Lafia	14 - 25	20 – 35	70 - 78	5.6 – 6.8	0.48 – 1.06
Akwanga	20 - 26	32 – 41	55 - 72	5. 6 – 6.2	0.88 – 1.26
N/Eggon	8 - 10	10 – 15	74 - 82	4. 8 – 6.8	0.46 – 0.68
Doma	28 - 45	18 – 25	50 - 65	4.5 – 6.4	0.68 – 1.88

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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 ± 5.92	24.0780 ± 2.92	408.6925 ± 56.14

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		17.4513 ± 0.27	46.055 ± 31.97	127.5869 ± 41.47

Table 4 Radiological Hazard Indices of Soil Samples Nasarawa State

Locations	Absorbed dose(nGy/h)	AEDR(uSv/h)x10^{-3}	Ra_{eq}(Bq/kg)	H_{ext}	$H_{int.}$	ELTCR (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	105.8801 ± 54.9	0.1299 ± 0.07	115.5043 ± 16.39	0.3120 ± 0.04	0.4022 ± 0.05	0.4547 ± 0.23

➢ *Annual Effective Dose Rate (AEDR)*

The AEDR was evaluated for the area, and it shows that the outdoor annual effective doses vary from 0.05 to 0.33 $uSvy^{-1}$, with the mean value of 0.13 $uSvy^{-1}$.In all the zones; the AEDE values exceed the World average value. These results showed a high annual effective dose compared to the World average value of 0.07$uSvy^{-1}$ for an outdoor effective dose. This could be due to the level of activity concentration of radionuclides in the Soils of the study areas and may be associated with the soils being rich in minerals. These values imply that the use of Soils from the farmland of the affected zones could pose a threat to bone marrow over time, leading to cancer. These may affect the people living or working in the area.

➢ *Radium Equivalent Activity (Ra_{eq})*

The Ra_{eq} values evaluated in this work, range from 81.77 to 159.09 $Bqkg^{-1}$, with an average value of 115.50 $Bqkg^{-1}$, which is below the safety limit of 370$Bqkg^{-1}$. The average value recorded in the entire zone is within the recommended limit. Hence, the Radium equivalent activities of all the zones do not pose any danger yet.

➢ *Hazard Index for Both External and Internal Indices*

The results were evaluated for both $H_{ext.}$ and $H_{int.}$, and were below unity (Table 6). These results presently show an insignificant radiation effect on farmers working in the farmlands of the areas and that the farmlands are appropriate for further use since most of the values obtained from the zone are less than one.

➢ *Excess Lifetime Cancer Risk (ELTCR)*

The values obtained for the ELTCR in the study area range from 0.172 x 10^{-3} to 1.159 x10^{-3} (Table 7). The average values obtained in the study area were above the world value of 0.229x10^{-3}(11). This implies areas where the values exceed the safety limit, the use of soils for farms and other activities in the area for a maximum of 70 years could pose a risk of cancer cases. Radionuclide keeps accumulating in humans via food ingestion and ambient radiations as the year goes by, and after so many years the effect will start manifesting in the body.

Table 5 Radiological Hazard Indices of Soil Samples in Nasarawa State

Locations	Absorbed dose(nGy/h)	AEDR(uSv/h)x10^{-3}	Ra_{eq}(Bq/kg)	H_{ext}	$H_{int.}$	ELTCR (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

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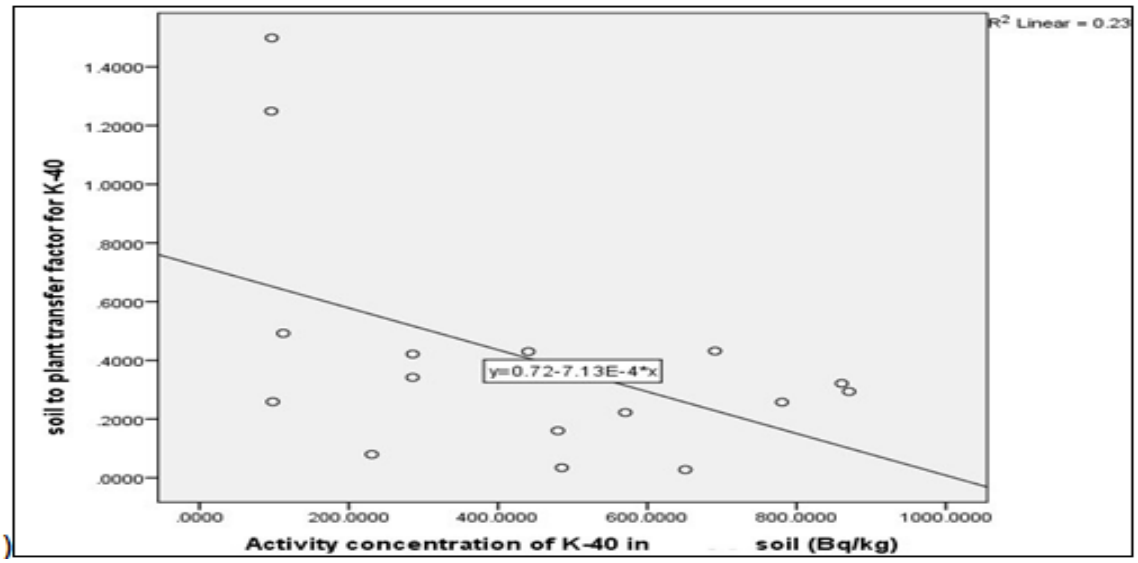
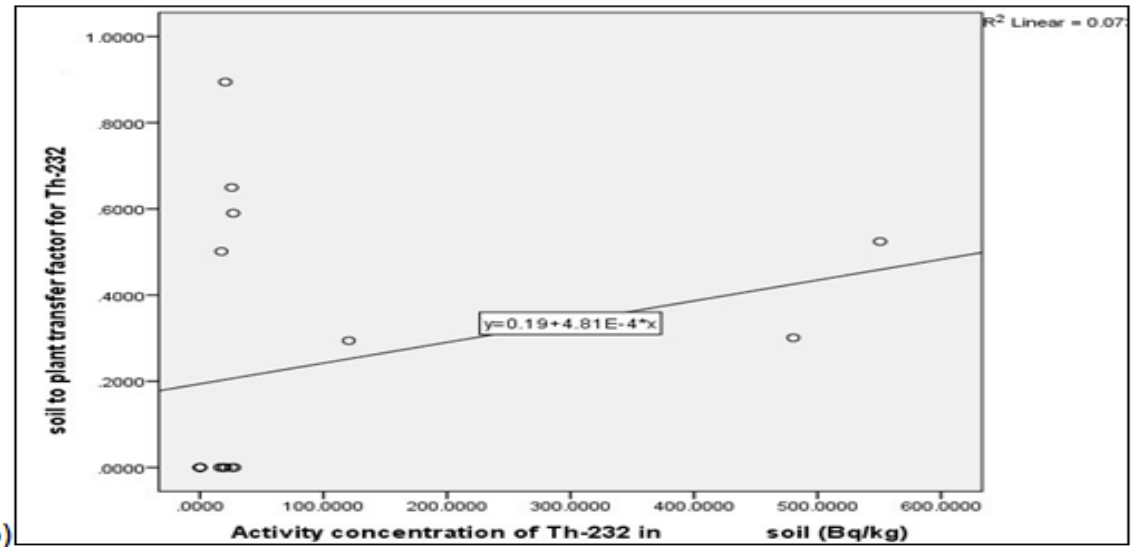
- *Correlation Analysis of Activity Concentration of Radionuclide in Soil and Crops and Linear Regression.*

Pearson's correlation was applied to analyze the concentration of radionuclide elements in soil and crop samples. The analysis performed showed that the activity concentration of ^{238}U in soil, and crop samples of the area was weak or had no correlation. Indicating the concentrations of radionuclide in crops does not depend on the activity concentration of the Soils in the area. Areas with a high activity concentration of ^{238}U in crops could have their concentrations from other sources like chemicals spread on crops or fertilizers applied directly on the root of crops or ambient radiations and not necessarily from the soil. On the other hand, the analysis for ^{232}Th shows a strong positive correlation (0.934) in the study area. These may be due to soil type and microbial activities in the soil. The correlation analysis performed for ^{40}K in the study area show a positive correlation between activity concentration of ^{40}K in soil and crop of 0.610 in the entire study area, meaning that there is a significant relationship between the activity concentration of ^{40}K in soil and activity concentration in the crop. Indicating an increase in activity concentration of ^{40}K in the soil leads to a corresponding increment in Crops. Potassium assists in regulating plant metabolism and also contributes to water pressure regulation inside and outside of plant cells. The high usage of NPK fertilizer in the farms could also account for a high concentration of ^{40}K in crops and soils. As earlier mentioned, the accumulation of ^{40}K in soil with corresponding accumulation in the crop may be affected by several factors such as cation exchange capacity (CEC), soil type, and soil pH.

Regression analyses were performed to compare the radionuclide activity concentration in Soils and the corresponding TF from soils to crops (Figure 2).The result shows is no linear relationship between the activity concentration of radionuclide deposits in soil and the TF. In some areas where there are high activity concentrations of radionuclides, the transfer factor of such places was low and vice versa. This means that the quantity of radionuclide in a location does not necessarily account for the transfer factor from soil to crop. Some factors like soil pH, cation exchange, organic matter, clay content, soil moisture, and soil type might have affected the presence and distribution of the radionuclides.



(a)



(b)

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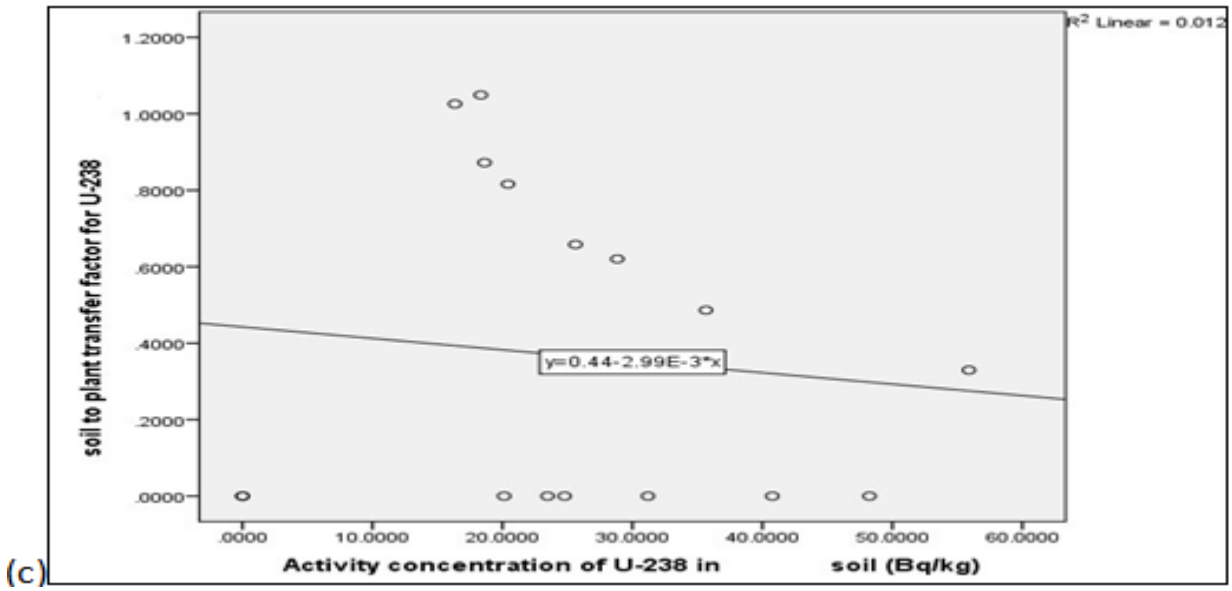
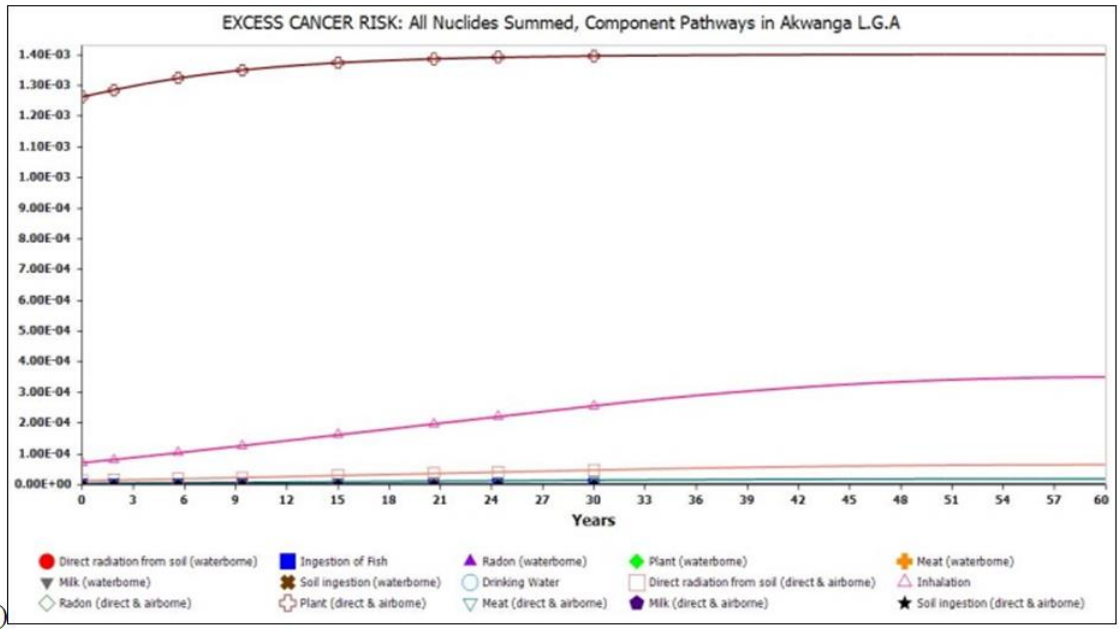


Fig 2 Regression Linearity Analyses of TF and Activity of Radionuclides (a, b and c)

➢ *Computation of Dose and Excess Lifetime Cancer Risk using RESRAD Offsite Model.*

The total maximum dose and excess cancer risk of the area were also evaluated to forecast future risk using the RESRAD-offsite model for individuals living in the study area. Figure 5-7 reviews the radionuclide dose and cancer risks for all the pathways summed for 60 years in the study area. The total highest radionuclide dose in 60 years for Akwanga was 1.03mSv/y at year 60, with thorium contributing more to the dose followed by potassium and Uranium last. The highest total cancer risk for all the pathways summed was 1.84 x 10^{-3}in 60 years. In Lafia, the total highest dose was 0.5 mSv/yr at 38 years, the highest total cancer risk for all pathways was 8.97 x 10^{-4}at 26 years, and Thorium is leading in the contamination here followed by potassium and Uranium last. In N/eggon, the total highest dose was 0.45 mSv/yr in 53 years and the highest total cancer risk for all pathways was7.28 x10^{-3} at 57 years, and in Doma, the total maximum dose was 1.08 mSv/yr at 51years, on the other hand, the total cancer risk for all summed pathway was 2.1 x 10^{-3} at 58 years.^{232}Th is driving the risk effect in the location followed by ^{40}K and then lastly ^{238}U. This is clearly illustrated in Figures (3). The world health organization's safety limit for cancer risk is 1×10^{-6} to 1×10^{-4} (13). This implies that the total cancer risks in the zone, particularly in Akwanga and Doma, were higher than the safety limits.

Considering the all summed component pathway analysis of the study area, it was discovered that the water-independent pathway has more impact in calculating the excess cancer risk in the entire zone. In this pathway, the cancer risk is largely caused by direct radiation from the crop (direct and airborne), which could be as a result of fertilizer application directly to the region of the crops, agrochemical applications on plants, and ambient radiation of the area.



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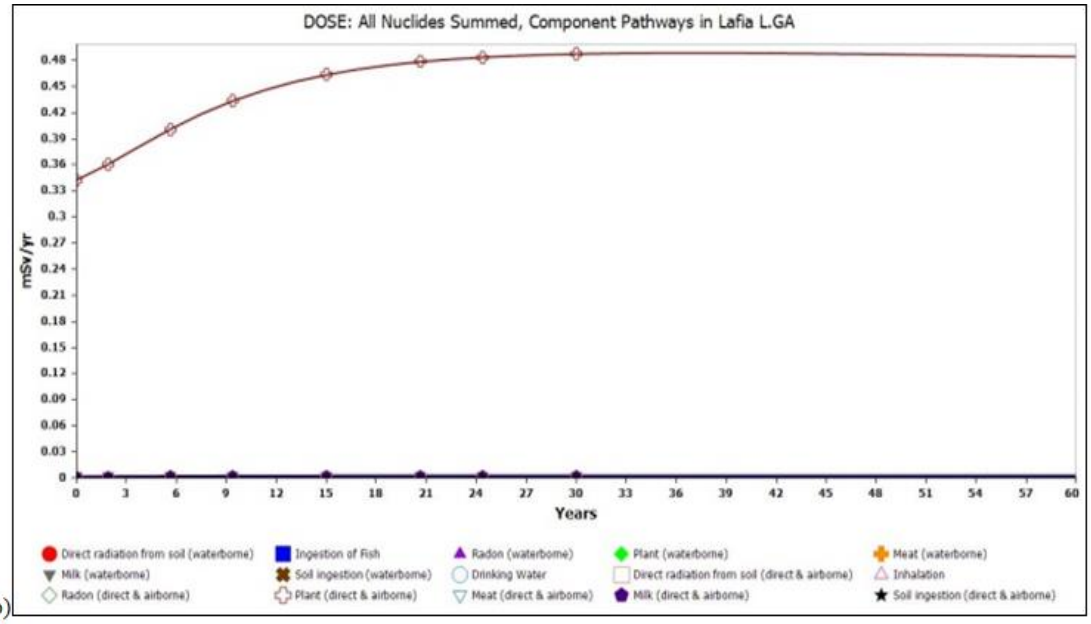

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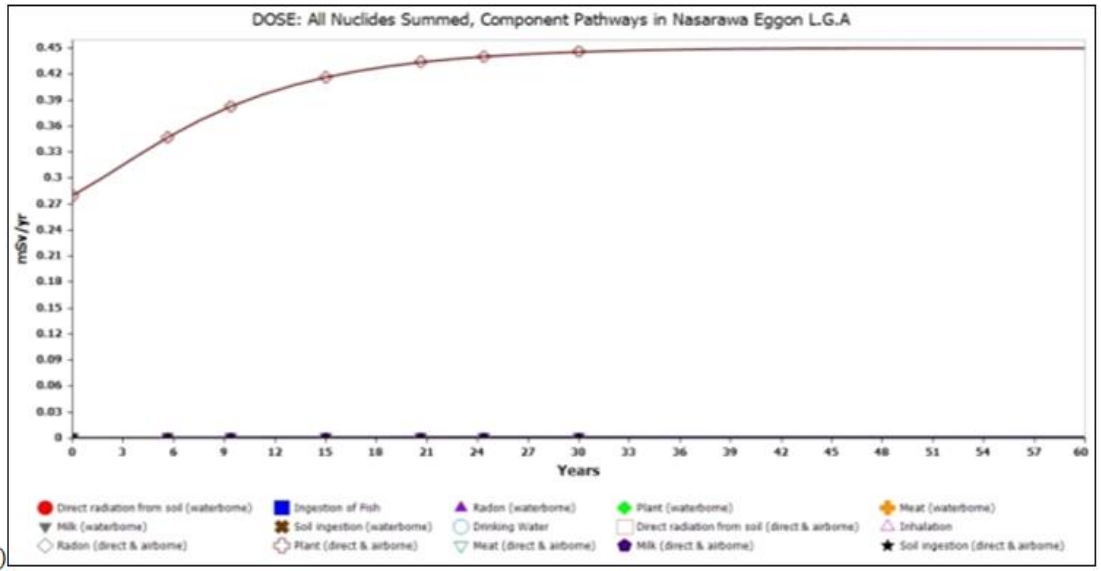

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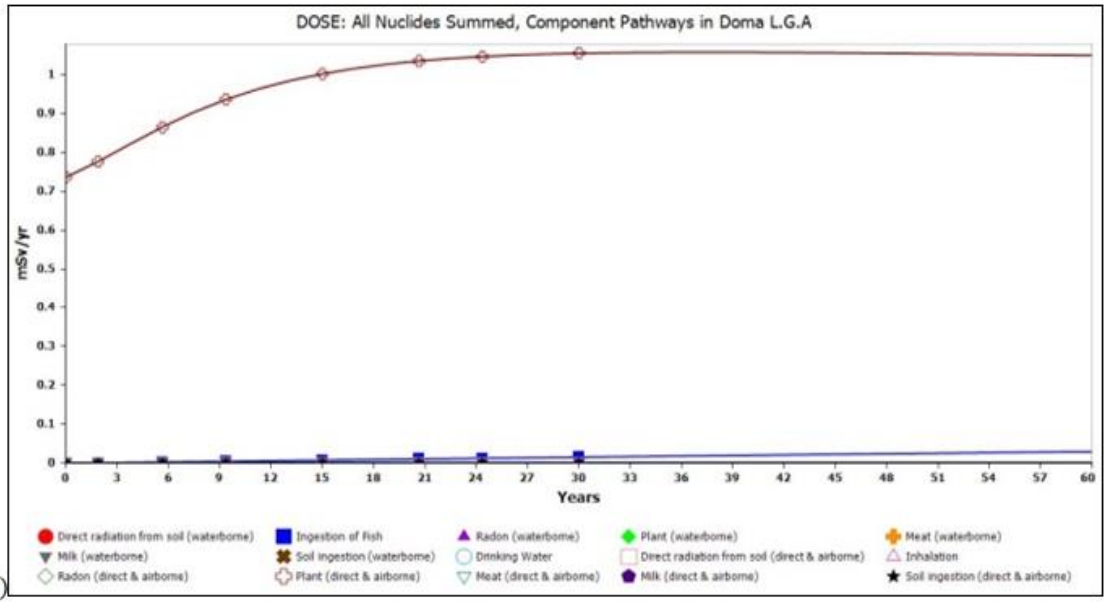

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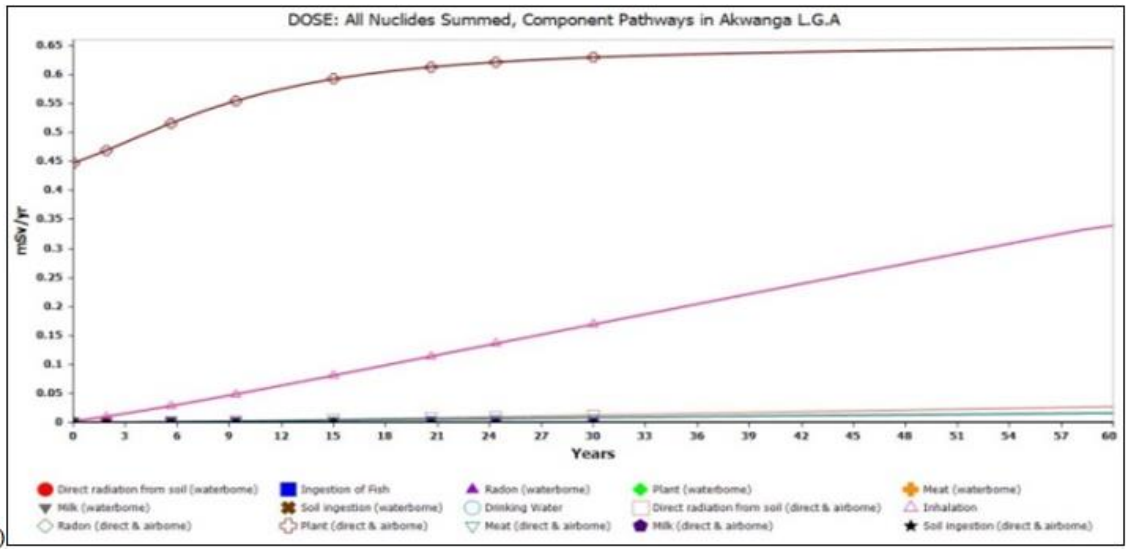
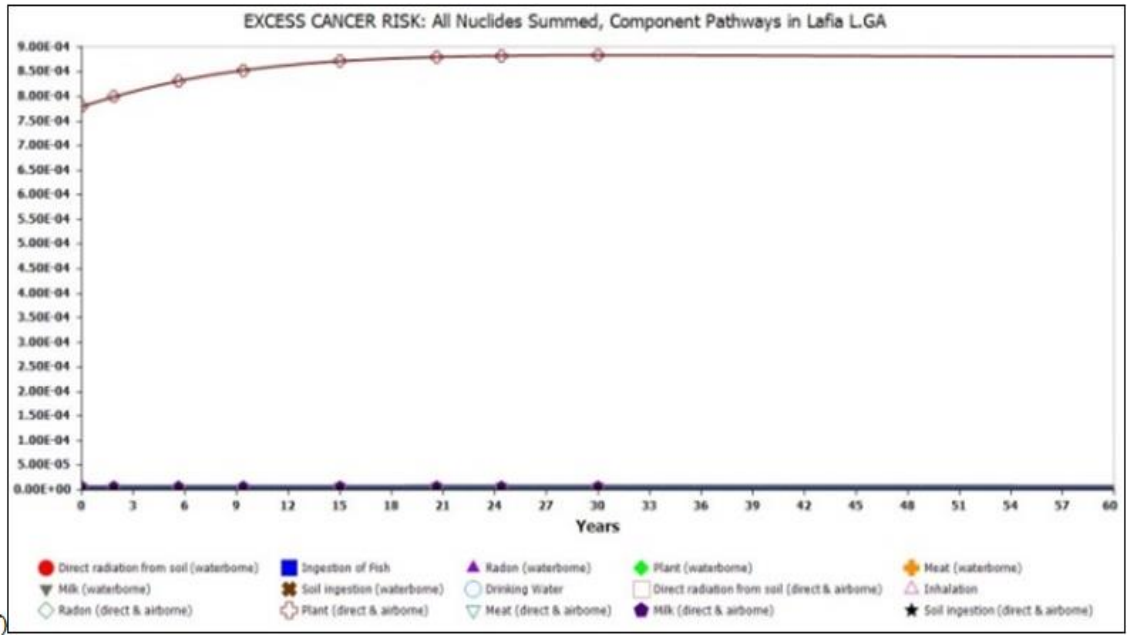
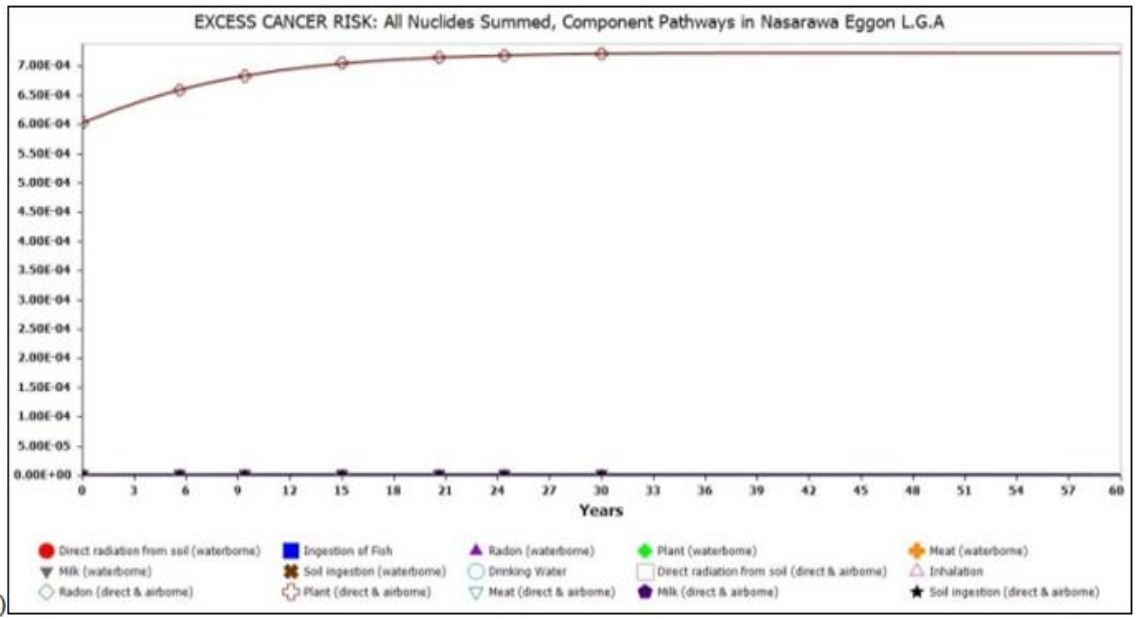

DOSE: All Nuclides Summed, Component Pathways in Akwanga L.G.A
mSv/yr
Years
Direct radiation from soil (waterborne)
Ingestion of Fish
Radon (waterborne)
Plant (waterborne)
Meat (waterborne)
Milk (waterborne)
Soil ingestion (waterborne)
Drinking Water
Direct radiation from soil (direct & airborne)
Inhalation
Radon (direct & airborne)
Plant (direct & airborne)
Meat (direct & airborne)
Milk (direct & airborne)
Soil ingestion (direct & airborne)

(e)


EXCESS CANCER RISK: All Nuclides Summed, Component Pathways in Lafia L.GA
Years
Direct radiation from soil (waterborne)
Ingestion of Fish
Radon (waterborne)
Plant (waterborne)
Meat (waterborne)
Milk (waterborne)
Soil ingestion (waterborne)
Drinking Water
Direct radiation from soil (direct & airborne)
Inhalation
Radon (direct & airborne)
Plant (direct & airborne)
Meat (direct & airborne)
Milk (direct & airborne)
Soil ingestion (direct & airborne)

(f)


EXCESS CANCER RISK: All Nuclides Summed, Component Pathways in Nasarawa Eggon L.G.A
Years
Direct radiation from soil (waterborne)
Ingestion of Fish
Radon (waterborne)
Plant (waterborne)
Meat (waterborne)
Milk (waterborne)
Soil ingestion (waterborne)
Drinking Water
Direct radiation from soil (direct & airborne)
Inhalation
Radon (direct & airborne)
Plant (direct & airborne)
Meat (direct & airborne)
Milk (direct & airborne)
Soil ingestion (direct & airborne)

(g)

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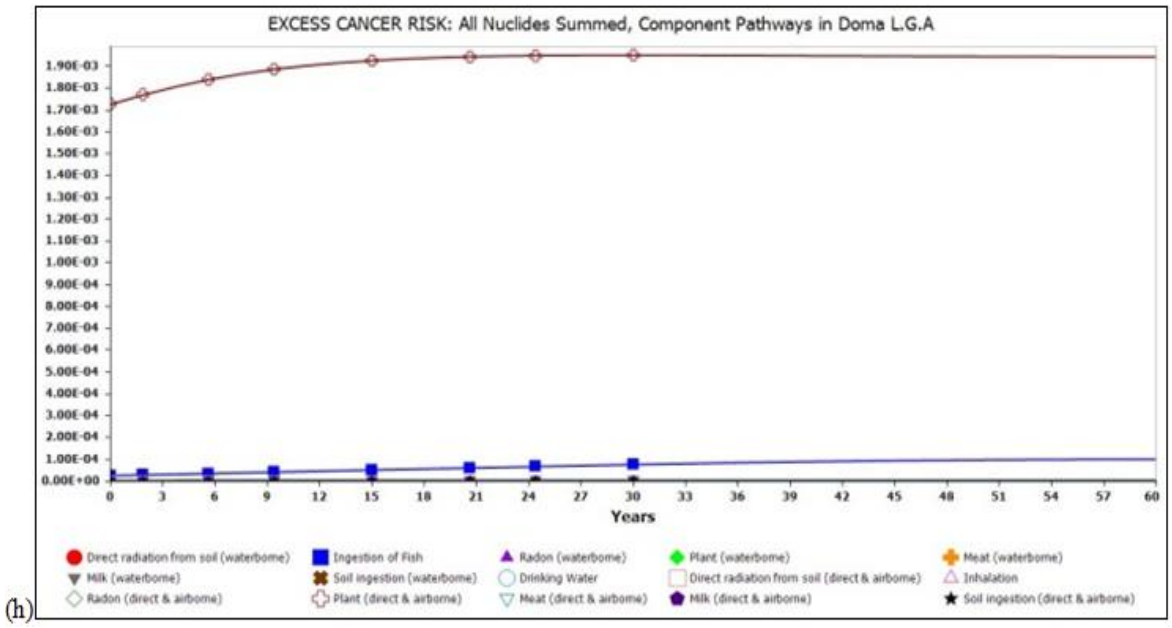

(h)

Fig 3 Dose Cancer Risk (a),(b), (c), (d) and Component Cancer Risk (e,f,g,h)

IV. CONCLUSION

The activity concentration in soil, and crop samples, and the TF of soil to the crop has been evaluated for 20 locations in Nasarawa state, Nigeria. The mean activity concentration 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, 46.06, and 17.45 Bqkg^{-1}, respectively. The Ra_{eq} concentration, the H_{ext}., and H_{int}. 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, 0.31, and 0.40Bqkg^{-1}, respectively. The average soil to crop TF for ^{40}K, ^{232}Th, and ^{238}U were 0.053, 0.369, and 0.366, respectively. The mean ADR and the mean AEDR in soil samples were 105.88 nGyh^{-1} and 0.13 mSvy^{-1}, respectively. The mean AEDR for the study area is higher than the world average (0.07mSvy^{-1}) and 0.1 mSvy^{-1} recommended by internal standards. The result available from this research work states that the activity concentration of radionuclides in crops is not solely a function of the number of radionuclide deposits in the soil. The summed component pathway analysis of the study area, reviewed that the water-independent pathway has more impact in calculating the excess cancer risk in the entire zone. In this pathway, the cancer risk is largely caused by direct radiation from the crop (direct and airborne), which could be as a result of fertilizer application directly to the region of the crops, agrochemical applications on plants, and ambient radiation of the area. The model also review that the total cancer risks in the zone, particularly in Akwanga and Doma, were higher than the safety limits by WHO.

- *Conflict of Interest*

The authors declare no conflict of interest.

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