

DETERMINATION OF ACTIVITY CONCENTRATION OF THORIUM-232 IN TOMATOES AND ESTIMATION OF ANNUAL EFFECTIVE DOSE TO THE PUBLIC IN NASARAWA L.G.A OF NASARAWA STATE, NIGERIA

AUDU, D. S.

Department of Science Laboratory Technology,
Federal Polytechnic Nasarawa.

E-mail: dsaudu4@gmail.com

Phone: +234-906-087-2112

Abstract

The purpose of this research work is to unveil the activity concentration of Th-232 in Bq/Kg in tomatoes (solanum Lycopersicum) and to determine the ingested doses of tomatoes consumed. A lot of ripped tomatoes were purchased from two different places in Nasarawa municipality, namely Tamma and Angwan Habibu and packaged into two big sacks and transported to the laboratory. The tomato samples were sliced and dried in open air under the radiation from the sun. Then the containers containing the tomato samples were allowed to stay for thirty (30) days for secular equilibrium to occur between the parent and the daughter radioisotopes. Measured activity that is counts/second (c/s) of the tomato samples were acquired at 1000volts for a period of 2hrs by using NaI (TI) detector (sodium iodide detector). Detector efficiency was determined by using standard point sources namely; ^{60}Co , ^{137}Cs , ^{22}Na and ^{54}Mn . Efficiency of Th-232 was found by method of interpolation from detector efficiency graph, for the purpose of calculating the activity concentration of Th-232 (Bq/Kg) in tomato samples. The activity concentration of Th-232 in tomato samples determined in the laboratory ranged from 16.73Bq/Kg to 17.32Bq/Kg. The estimated dose value due to tomato samples consumed is 3.84 Sv/yr to 4.01Sv/yr in Nasarawa local government area of Nasarawa State. This estimated doses due to ingestion of tomatoes food samples is below the safe limit recommended by UNSEAR which is 290Sv/yr. Thus, there is no any health hazard as result of consumption of these tomato samples within the environment.

Keywords: Activity Concentration (AC), Tomatoes (solanum Lycopersicum), Radiations, Sodium Iodide detector, Annual effective dose.

Introduction

Vegetation in Nasarawa L.G.A is mostly grassland, trees and herbs usually scattered. There are two season here that is, dry season and wet season. During the dry season grasses usually die and in the wet season the grasses come back. Nasarawa town is found within longitude 7° and 8°E and latitude 8° and 9°North belonging to southern guinea savannah of annual rainfall of 1000-1200mm.

People in this region do farming on cassava, yam, sweet potatoes, tomatoes, pepper, millet, maize, and a host of others (Audu, 2000). Natural resources like Fe, tin, zinc, Mn, columbite, tantalite are found within the Nasarawa L.G.A. Also present within the environment are primordial radionuclides from the earth crust of various activity levels which external gamma radiation arises. These radionuclides comprise of Th-232 decay series, U-238 decay series and K-40. The percentage contribution of these radionuclides in the environment is about 99% radiation (Guy, 1999). There are different concentrations of micro-elements and radioisotopes present in various segment of air, water, food, soil and plants which has been recorded over the years (Andras, 1993).

Recent research work on tomato in Jos metropolis of Nigeria (Fom et al, 2015) indicated the presence of Cu in concentration of 150 and 42mgdm^{-3} respectively in Naraguta and Demili village fadama. Similarly Cd concentration in Naraguta village was 3mgdm^{-3} above the safety

limit of 0.8mgdm^{-3} (Fom et al, 2015). Zinc concentration at Bauchi Bridge Fadama recorded 192mg/dm^3 which is below the safe limit of 200mg/dm^3 (Fom et al, 2015). Also zinc concentration at Naraguta Village Fadama and Demili Village Fadama recorded 300mg/dm^3 which was above the safe limit of 200mgdm^{-3} (Fom et al, 2015). The concentrations of these metals were high in tomatoes in some areas because of application of manure, artificial fertilizer on the farmlands. And also the presence of metals in the soil particles displaced through water erosion that were absorbed by tomatoes (Fom et al, 2015).

Also in the region of south Kordofan state (Sudan) other researcher's result of Th-232 varied from $2.40\text{Bq/Kg} - 5.15\text{Bq/Kg}$ by Hatem Eltayeh Fadlalla Hemada (2009), with effective dose as 1.49mSv/y which is below the safe limit of 1mSv/y for the general public. In Red Sea state (Tokor), the activity concentration varies from $(2.57 - 2.57\text{Bq/Kg})$, with estimated dose of 1.21mSv/y , $1.55 - 7.35\text{Bq/Kg}$ at River Nile with estimated dose of 3.05mSv/y , $(0.03 - 3.15)\text{Bq/Kg}$ was determined in the northern part of Sudan with estimated dose of 1.25mSv/y (Hatem Ectayeb Fadlalla Hemada, 2009). The result of Th-232 AC in tomatoes recorded in Port Harcourt are as follows; 105Bq/Kg with estimated dose of 24.31mSv/y (Ononugbo et al, 2017). In the same vein, other results of Th-232 in tomato are as follows in terms of estimated doses are 5.63 in Nankhan and 64.92 in Cloves all in Saudi Arabia (Al-Graham, 2014 as cited in Ononugbo et al, 2017).

Tomatoes are well consumed but less work has been done on it. Most researchers focused on other food spices or samples like pepper, onion, garlic, melon seed, ginger and a host of others with their respective activity concentration (Bq/Kg) as follows; 6.09, 40.57, 5.95 and 7.2 were determined (Ononugbo et al, 2007). So, it becomes very interesting to access or study the activity level of Th-232 in tomatoes and its ingestion doses to the public within Nasarawa L.G.A of Nasarawa State.

The aim of this research work is to measure the activity concentration of naturally occurring Th-232 in tomato samples via gamma ray spectroscopy and hence estimate annual effective dose.

AC is the activity concentration expressed in Bg/kg i.e. Activity concentration (AC) = Bq/Kg ----- (1) (Audu, 2000), (Amin et al, 2013), Ononugbo et al, (2017).

Where c/s = counts/second in the sample (measured activity)

Th_ϵ = efficiency of Th-232 obtained from detector efficiency graph by method of interpolation. B.R. = branching ratio of Th-232 and m^1 = mass of the sample in kg, i.e $m^1 =$.

And Estimated annual effective dose is given as $E = A_t \times I_s \times \text{DCF}$ (Ononugbo et al, 2017). Where E = annual effective dose by ingestion of the radionuclide in the sample (Sv/y), A_t = Activity concentration in the sample (Bq/Kg), I_s = Annual intake of food spices (kg/y) as 1kg/y , DCF = Internal dose conversion factor by the ingestion of the radionuclide (Sv/Bq) given as 0.23Sv/Bq for Th-232 (UNSCEAR, 1993) as cited in Ononugbo et al, 2017).

The choice of Nasarawa L.G.A for the research work was chosen because the author works at Federal Poly. Nasarawa. Secondly the presence of, tantalite, zinc, Fe, U, k-40, Th-232 etc are within the soil (Audu, 2000). Thirdly mining activities of these natural resources have been carried out at Tamma and A/Habibu via open pit mining by Squilos Nig. Ltd in the year 1989/1999 etc from record available in Nasarawa L.G.A, 1999 (Audu, 2000). Thus the presence of Th-232 and their mineral resources in these areas stimulated this research finding in tomato samples.

Many researchers focused their attention on food spices or samples like pepper, onion, garlic etc and doing less work on tomatoes equally necessitated this research work.

Thorium and their compounds

Thorium is a radioactive element that occurs naturally in low concentrations about 12ppm in the earth crust. Thorium in its pure state is a white silvery heavy metal that is as dense as lead. Besides Th-232, there are other isotopes of Thorium that exist in nature in small amount (isotopes are different forms of an element that have the same number of protons in the nucleus but a different number of neutrons). Thorium is soft, ductile metal pyrophoric in powdered form. When heated in air thorium turnings ignite and burn brightly with a white light (Leiterer et al, 2010). The main source of Thorium is monazite sands. The compounds of Thorium are Thorium Oxide (ThO_2), Thorium silicate (ThSiO_4), Thorium Phosphate (ThPO_4). The concentration of thorium oxide in monazite sand is about 3 to 10%. Large resources are found in Australia, Brazil, U.S.A, Canada, India, South Africa and other countries including Nigeria of 90,000tons of thorium equivalent (Leiterer et al, 2010).

Tomato plants absorb thorium ions from the soil solution or soil colloids by diffusion method (Dosekun, 1980).

Th-232 has a natural abundance of 100% and designated in activities series as $4n + 2$ and is the leader of decay series which emits alpha, beta (particles) and gamma radiations and reaches a stable element Pb-208 (Keifer, 1990).

Uses of Thorium and their compounds

Thorium is used as gas lamp mantles, in ceramics to make them heat resistant.

It is used as an alloying element with magnesium, used as fuel for nuclear reactor, coating of tungsten wire for component of electronic equipment. Also thorium oxides are used to make diagnostic x-ray photographic in the hospitals e.g Th-232 dioxide, thorotrast (R), and also electronic bulb, bulb filament, welding rods and a host of others (Leiterer et al, 2010).

Route of Th-232 into the body organs.

Thorium (Th-232) concentration in the environmental samples e.g pepper, tomatoes, garlic etc varies according to geographical and geological factors.

Thorium can be taken into the body of man and animals by eating food, drinking water, or inhalation. Most of the thorium that is inhaled or ingested through food and water is excreted within some days from blood stream depending on its biological half-life. Of this amount entering the blood, some are deposited in the bone, liver, lungs, kidney and so on and the rest are uniformly distributed to other organs or tissues of the body. (Amin et al, 2013)

Side effects of Thorium

Thorium is generally health hazard only if it is taken into the body. Deposition of Th-232 in large quantities in a particular organs produced radiation damages, biochemical and morphological changes which result in weakening of immune systems, causes various type of diseases e. g cancers (brain cancer, bone cancer) and a host of others and increase the mortality rate (Tykua & Sabol, 1995 as cited in Amin et al, 2013).

Tomato (*Solanum Lycopersicum*): Tomato fruits are succulent in nature and is an example of berry which is widely consumed as food spices in Nigeria and other parts of the world with their seeds embedded in the endosperm. The epicarp, and the mesocarp of tomatoes are redish in colour. Besides Th-232, present in tomatoes there are other components present in tomatoes like fibre content, vitamin C, vitamin A (carotenoid), lycopene and potassium (which has nutritional value) (Freeman et al, 2010).

Uses of Components Present in Tomato

Lycopene is the major carotenoid found in tomatoes which is responsible for the red-deep colour of ripped tomatoes while vitamin A is very useful to the body as it promotes healthy growth and makes the eyes to function properly and helps the body to pose high resistances to diseases. For vitamin C it maintains the strength of blood vessels, cholesterol reduction in the blood and prevent mouth scurvy. Nutrient like potassium contributes to blood pressure regulation and helps to manufacture their proteins, carbohydrate etc and are incorporated into the endosperm of the fruit for their healthy growth.

Fibre content is for lowering cholesterol in the blood and regulate blood glucose concentration. In addition to that, for skin protection, tomato intake (40g of tomato paste corresponds to lycopene dose of 16mg) approximately for more than 8 weeks reduced ultraviolet light induced skin damage e.g erythema (Stahl et al, 2001as cited in Freeman et al, 2010).

Statement of the problem are cost and availability of tomatoes, the agricultural factors like good soil and vegetation, no enough rainfall could inhibit the availability of tomatoes for research work.

The Equipment (Sodium Iodide Detector or Scintillation counter)

Sodium Iodide detector [NaI (TI)] is the most commonly used scintillation counter for gamma ray spectroscopy. The detector has a diameter of 0.75m and thickness of 0.25m with its relative density of $3.67 \times 10^3 \text{kg/m}^3$ and high atomic number. Has 100% intrinsic detector efficiency, decay constant of 0.25s and dead time of 1 – 5s. The daughter radioisotope i.e Thallium (TI) in the sodium iodide detector is an activator or impurity which is responsible for the luminescence of the crystal. The detector is brittle and hygroscopic in nature.

The reason for [NaI(TI)] being used as Scintillation Counter

The emission of the scintillation counter picks at 410nm at the energy of 1400keV. And at this energy, radiation emitted from the sample source are measured by scintillation counter as count/seconds and register as peaks (Nicholas, 1983). The energy emitted from the sample source, the time particle is arriving at the counter, and the number of the particles emitted by the sample are used as a tool to measure the radioactivity of the samples and this activity aided to calculate the concentration of any radionuclide present in the sample. The scintillator are crystals that give out sparks or light photons when ionizing radiation passes through them. The scintillation counter is composed of detector housing, photomultiplier tube and amplification unit. The operation of the scintillation counter involves absorption of incident radiation energy, amplification of the light photons to give out pulse as peaks (Nicholas, 1983).

Materials and Methods

Collection and treatment of Sample

Ripped tomatoes (*Solanum lycopersicum*) were purchased from retailers in two different areas, Tamma and Angwan Habibu within Nasarawa municipality of Nasarawa L.G.A. and packed into two big sacks and transported to the laboratory. The tomatoes samples were sliced into pieces and dried under the influence of radiant energy from the sun for ten (10) days, spread on dried cleaned trays. Weighed quantities were packed into ten (10) dried cleaned plastic container (13.5cm in height and 5.6cm in diameter) sealed and tightly covered to avoid leakage of daughter radioisotope. Then the containers were allowed to stay

for 30 days so that secular equilibrium will occur between the parent and daughter radioisotope and the samples are ready prior to instrumental determination.

Experimental Work

Instrumental determination of activity concentration

Different weighed quantities of tomato samples (seed/pericarp) e.g (0.120kg, 0.121kg etc in Table 1) already inside the plastic containers were dropped one after the other into the detector well [NaI(Tl) detector]. The detector is connected to multichannel analyzer (MCA) coupled to a computer used for gamma ray spectroscopy at 1000V. Each sample was counted for 2hrs (duration or exposure time) and recording counts/sec for each sample until all the radiation from the tomato samples were acquired. The detector efficiency was determined by using standard points sources and this enable the identification and quantification of the radionuclides present in the sample using IAEA standard, 2000 (Onunogbo et al, 2017). The points sources are as follows ^{60}Co at energy (1172 and 1331)keV, ^{137}Cs at energy (662.2)keV, ^{22}Na at energy (508.4 and 1279.47)keV and ^{54}Mn at energy (833.4keV).

The detector efficiency obtained in this research work is in line with that report by Knoll, (2000). Because of this fact, NaI(Tl) detector was used in this research to determine the concentration of Th-232 in Bq/Kg in tomatoes sample at 2658.5Kev line. Initially the background radiation was acquired and stored in the computer and later on the background counts (measured activity) was subtracted from the sample counts to give the actual counts of the samples.

The activity concentration is calculated using equation (1). The efficiency of Th-232 was obtained from the graph of efficiency versus energy (point sources) which is 2.5% by method of interpolation.

$$\text{Activity concentration (AC)} = \text{Bq/Kg} \text{ ----- (1)}$$

Where c/s = counts/second in the sample (measured activity)

Th_ϵ = efficiency of Th-232 obtained from detector efficiency graph by method of interpolation. B.R. = branching ratio and m^1 = mass of the sample in kg, i.e $m^1 =$

Estimated Annual effective doses of tomatoes due to ingestion into the body is calculated from the equation below.

$$E = A_t \times I_s \times \text{DCF} \quad (\text{Ononugbo et al, 2017}).$$

Equation is rewritten as;

$$E_{\text{Th-232}} = A_{\text{Th-232}} \times I_s \times \text{DCF}_{\text{Th-232}} \text{ -----}^{(5)} \text{ and applied in the calculation for estimated dose to the body.}$$

Where E = annual effective dose, equivalent by ingestion of the radionuclide in the sample (Sv/y), A_t = Activity concentration in the sample (Bq/Kg), I_s = Annual intake of food spices (kg/y) as 1kg/y, DCF = Internal dose conversion factor by the ingestion of the radionuclide (Sv/Bq) given as 0.23Sv/Bq for Th-232 (UNSCEAR, 1993) as cited in Ononugbo et al, 2017.

Result and Discussion

Table 1 shows the concentration of Th-232 in the collected tomatoes from two locations in Nasarawa L.G.A.

Table 1: Th-232 Concentration in Tomatoes Samples at 2658.8keV

S/N	Sample Code	Location	Mass(Kg)	Annual Effective Dose (Sv/y)	Measured Activity (c/s)	Activity Conc.
1.	Tt.1	Tamma	0.120	0.0311	16.63	3.82
2.	Tt.2	"	0.122	0.0313	16.73	3.84
3.	Tt.3	"	0.121	0.0315	16.84	3.87
4.	Tt.4	"	0.236	0.0316	16.89	3.91
5.	Tt.5	"	0.125	0.0318	16.99	3.91
6.	Tt.6	A/Habibu	0.127	0.0319	17.10	3.93
7.	Tt.7	"	0.127	0.0320	17.11	3.94
8.	Tt.8	"	0.130	0.0321	17.16	3.95
9.	Tt.9	"	0.134	0.0322	17.21	3.96
10.	Tt.10	"	0.132	0.0324	17.32	4.01

Discussion

The activity concentration (AC) of Th-232 in tomatoes samples from Nasarawa L.G.A. as determined in the laboratory varies from 16.73Bq/Kg to 17.32Bq/Kg.

The concentration of Th-232 in the tomatoes from A/Habibu in Tt.6 was high and range from (17.10Bq/Kg) to 17.32 Bq/Kg with the estimation dose of 3.93Sv/y – 4.01Sv/y due to tomatoes ingestion.

The result obtained at Angwan Habibu ranged from 17.10Bq/Kg (Tt.6) to 17.32Bq/Kg (Tt.10) almost agreed to what was obtained in the literature as 18.7Bq/Kg in tomatoes sample (Amin et al, 2013). The estimated dose of the tomatoes samples after intake is 4.3Sv/y recorded by Amin and Ahmad, (2013), through gamma rays spectroscopy. Furthermore, one of the result of Th-232 in tomatoes sample as cited in the literature is 17.33Bq/Kg using gamma ray spectroscopy (Onougbo et al, 2013) and this result 17.33Bq/Kg in the literature agreed with the one obtained from the research work, i.e. 17.32Bq/Kg (Tt.10) at Angwan Habibu (A/Habibu) with estimated dose of 4.01Sv/y for tomatoes consumed by the populace.

Similarly, at Tamma, activity concentration (AC) of Th-232 in tomatoes ranged from 16.63Bq/Kg (Tt.1) – 16.99Bq/Kg(Tt.5) with estimated dose of 3.91Sv/y. In this area activity concentration in tomatoes samples was below what was found at A/Habibu all in Nasarawa L.G.A of Nasarawa State.

The activity concentration of Th-232 in tomatoes sample from the research work does not exceed the 1000Bq/Kg (IAEA, 1989) as the permissible limit for gamma emitters in food. The result of annual effective dose (4.01Sv/y) received by individual ingestion of food spices (tomatoes) is lower than the world safe limit of 290Sv/y for annual intake of One(1) Kg/yr for all samples food spices UNSCEAR, (2000) as cited in Ononugbo et al, (2017). According to Amin et al, (2013), annual effective ingestion doses due to intake of food spices is due to the scale of annual intake of 1Kg/yr.

Furthermore, the justification of the differences in the level of Th-232 in this study is due to the variation of the concentration of Th-232 in the soil at A/Habibu and that of Tamma, and also is because of geological factors which include soil composition and a host of others from Audu (2000). The mineral deposits and radionuclide concentration in the soil at A/Habibu is higher than that of Tamma via gamma ray spectroscopy (Audu, 2000). It implies that tomatoes from A/Habibu will have more Th-232 to absorb than the ones from Tamma. Similarly, leaching (water drainage) is another added factor which is higher at Tamma than at A/Habibu. So the soil here is sandy (Audu, 2000). By leaching radionuclides are being displaced from one place to the other (Audu, 2000), which lead to the depletion of Th-232 in tomatoes in Tamma.

The author's result in this research work agreed with the findings of some previous result as cited in the literature showed that Th-232 contained in tomatoes from Nasarawa L.G.A have almost the same concentration with that of Amin et al, (2013) in Egypt. Thus the authors work is consistent and reliable.

Conclusion

The activity concentration (AC) in Bq/Kg of Th-232 in tomatoes (*solanum lyco periscum*) in Nasarawa municipality (A/Habibu and Tamma) all in Nasarawa L.G.A of Nasarawa State varies from 16.73 Bq/Kg - 17.32 Bq/Kg with estimated dose due to ingestion of food spices (tomatoes) as 3.82 μ Sv/y - 4.01 μ Sv/y. The activity concentration of tomatoes sample determined in the lab. by gamma ray spectroscopy does not exceed the safe limit of IAEA, 1000 Bq/Kg for gamma radiation emitters in food. Also, determined is the annual effective dose of 4.01 μ Sv/y received by the public due to in takes of food spices (tomatoes) is lower than the recommended value of 290 μ Sv/y.

Thus tomatoes samples analysed in this research work are radiologically safe for human consumption as per the international standards. So, diseases like brain cancer, bone cancer, and other related diseases as cited in the literature were not found among the populace of Nasarawa L.G.A. This result could be used as a reference frame in terms of radiological standard on food spices for future studies within the locality. More so, a lot of precautions must be taken in eating any food such that the dose limit must not be exceeded since we don't know how much radionuclides are present in our bodies. To keep the equipment [NaI(Tl) - detector] encapsulated when it is not in use is okay because the equipment is hydroscopic.

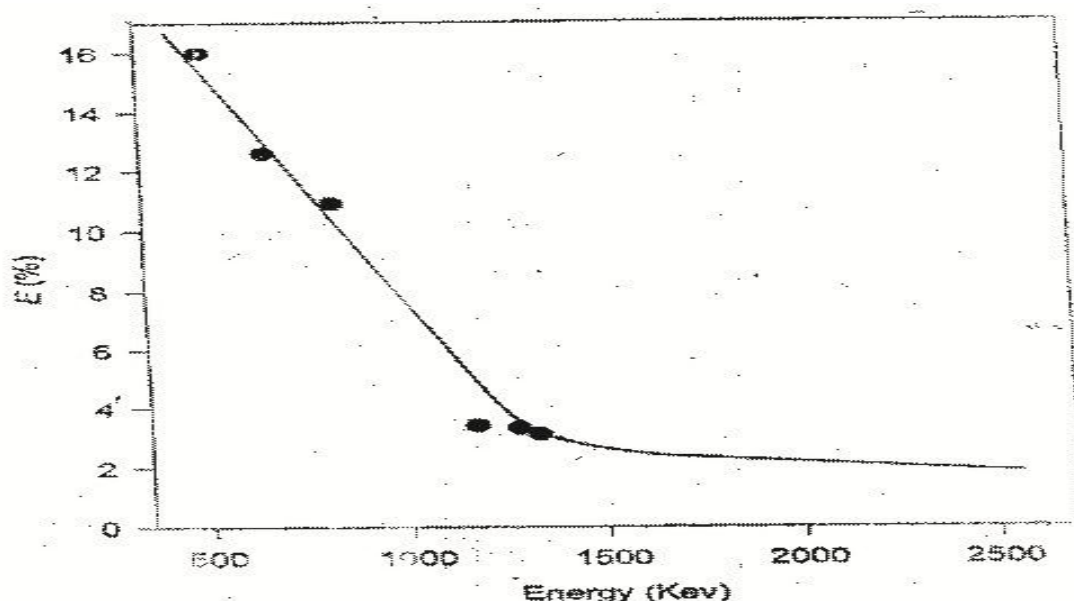


Figure 1: A graph of efficiency Vs Energy (point sources) Showing Sodium Iodide detector efficiency

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Appendix

To calculate the activity in Becquerel (Bq) which is the number of disintegration per second (d/s). One must know the counting efficiency which **is** defined as the ratio of observed counts per second to the disintegration per second. Counting effecting = (c/s) x 100% ----
(1) (1) is rewritten as $A_t = \text{Bq}$ ---- (2). In terms of thorium (Th-232) equation (2) becomes $A_{\text{Th-232}} = \text{Bq}$. In terms of activity concentration (2) becomes,

$$AC_{\text{Th}} = \text{Bq/Kg} \text{ -----(3)}$$

(Audu, 2000), (Amin et al, 2013), (Ononugbo et al, 2017)

Where A_t is the activity at time t which is taken as the activity of Th-232.

Efficiency of Th-232, obtained from interpolation of efficiency graph (curve) as 2.5%. Bq Background radiation counts (c/s) subtracted from sample counts B.R Branching ratio 99.8% for Th-232 and m^1 = mass in Kg (mass/kilogram).

Estimated Annual effective doses of tomatoes due to ingestion into the body is calculated from the equation below.

$$E = A_t \times I_s \times \text{DCF} \quad (\text{Ononugbo et al, 2017}).$$

The above equation is rewritten as;

$$E_{\text{Th-232}} = A_{\text{Th-232}} \times I_s \times \text{DCF}_{\text{Th-232}} \text{ -----(5)} \text{ and applied in the calculation for estimated dose to the body.}$$

Where E= annual effective dose by ingestion of the radionuclide in the sample (Sv/y), A_s = Activity concentration in the sample (Bq/Kg), I_s = Annual intake of food spices (kg/y) as 1kg/y, DCF = Internal dose conversion factor by the ingestion of the radionuclide (Sv/Bq) given as 0.23Sv/Bq for Th-232 (UNSCEAR, 1993) as cited in Ononugbo et al (2017).