



Assessment of Natural Radionuclide Ingestion through Foodstuffs Cultivated Around the Sokoto Cement Production Facility, Northwestern Nigeria

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ABSTRACT

Exposure to radiation is a natural phenomenon unavoidable to human environment. Ionizing radiation is received from naturally occurring radionuclides in water, air, soil and food. Ingestion is one of the major pathways through which these radionuclides enter the human body. Assessment of natural radionuclide on twelve (12) samples, four (4) each of leafy vegetables, cereals and tuber crops cultivated and consumed around the cement production facility of Sokoto has been carried out using a 76mm by 76mm lead-shielded Sodium Iodide detector (NaI(Tl)) located at the Centre for Energy, Research and Training (CERT), Ahmadu Bello University Zaria, Nigeria. The overall mean activity concentration of the radionuclides in the samples of leafy vegetables, cereals and tuber crops were 39.8904 ± 6.1347 Bqkg⁻¹ for ²²⁶Ra, 57.6631 ± 5.5907 Bqkg⁻¹ for ²³²Th and Bqkg⁻¹ for 168.6004 ± 8.4195 ⁴⁰K. All values of consequential effective dose for both adults and children due to consumption of radionuclides in food stuffs were higher than recommended value of 1 mSvyr⁻¹, except for leafy vegetable in children. Hence, intermittent monitoring of radioactivity level in food stuffs grown in the study area is recommended.

Keywords: Natural Radioactivity, Effective Dose, Food Stuffs, Gamma Spectrometry, Sokoto Cement Company

INTRODUCTION

Environmental quality perturbation resulting from the increase in the level of potentially toxic substances is becoming more pronounced, thus raising the question on the safety status of the environment. Industrial activities rank higher among contributors to environmental degradation (Prince *et al.*, 2020). Non-nuclear industries, such as cement industry, use materials containing significant contents of naturally-occurring radioactive materials (NORMs), which may expose workers or the people living near such plants to radiation above the natural background (Adekoya *et al.*, 2022). These radionuclides have been essential constituents of the earth, since its creation (UNSCEAR, 2000). The abundant presence of naturally occurring radioactive materials (NORMs) is very hazardous to humans' health (IAEA, 2015). The main contributors of natural radiation in the environment are the high energy cosmic radiations that are produced constantly in the atmosphere; and the primordial radionuclides of the uranium (²³⁸U) with a half-life of 4.5×10^9 years and thorium (²³²Th) decay half-life 1.405×10^{10} years series as well as radioactive potassium (⁴⁰K) with a half-life of 1.277×10^9 years in naturally occurring radioactive materials (NORM) existing in the earth crust since the formation of the earth known as radionuclides (Pourimani *et al.* 2015, ; Issam *et al.* 2018). These radionuclides attract attention because of their high instability, which calls for their long decay series in order to achieve stability (Ocheje & Tyovenda, 2020). The inhomogeneous distribution of these radionuclides in geological formations (soils and rocks) however suggests that

man made products derived from substances (such as cement) will contain traces of these radionuclides whose concentrations will be dictated by the origin of the soil and rocks they are derived from (Ibrahim, 2015).

Foodstuffs are also known to be embedded with different proportions of radionuclide content depending on where they are cultivated (Fasanmi *et al.*, 2021). Moreover, inhabitants of communities near mining activities such as those of cement production are heightened also depend on the farm produce cultivated in their area for sustenance. The two major pathways by which natural radionuclides enter into man are ingestion and inhalation of different sources of food (Tawalbeh *et al.*, 2012). Cultivation of crops meant for consumption in contaminated environment result in the transfer of radioactivity from soil to the roots of such crops, which is consequently shifted to human diet (Fasanmi *et al.*, 2020). Doses of exposure to radiation by ingestion are due mainly to the presence of ²²⁶Ra, ²³²Th, and ⁴⁰K radionuclide series that are found in food and drinking water (Chen *et al.*, 2016). However, feeding on food items that have a high content of radioactive contamination can result in adverse health consequences such as respiratory issues or cancer (Hamada & Ogino, 2012). Studies have shown that undue exposure to radiation over a prolonged period can damage the organs of the body when it is beyond the recommended safe limit (Jibiri & Okusanya, 2008).

This ingested radionuclide could be concentrated in certain parts of the body thereby causing tissue damage as shown in Figure (1) (IAEA, 1989).

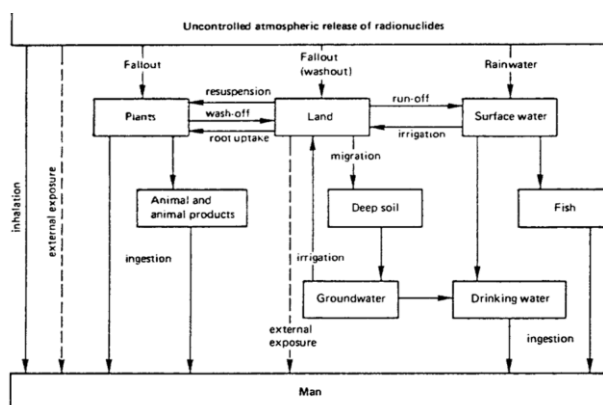


Figure 1: Major Pathways of Radionuclides to Man In The Event Of an Uncontrolled Release of Radioactivity (IAEA, 1989)

Studies regarding the analysis of natural radioactivity in foodstuff have been pushed to the forefront in recent years, and it has become an important aspect of the environmental monitoring program. Food stuff is one of the major pathways through which radionuclide is transferred to man.

It is therefore necessary to estimate the radiation doses obtained from the ingestion of contaminated food (Salim *et al.*, 2021; Syarbaini *et al.*, 2014). Several studies have been conducted to measure and determine radionuclide concentrations and resultant doses in different sources of food in Nigeria and beyond (Irunwor *et al.*, 2022; Fasanmi *et al.*, 2021; Essien, *et al.*, 2021; Mohammed *et al.* 2018).

Cassava tubers cultivated in the Ewekoro cement area, Southwestern Nigeria were investigated with the aid of Hyper Pure Germanium (HPGe) detector in order to assess the radionuclide content. Samples of cassava reported the highest activity concentrations of 228.15, 79.26 and 81.50 Bqkg⁻¹ for ⁴⁰K, ²³²Th and ²²⁶Ra, respectively. All the above results were higher than the recommended safe limits by a factor of 2. Moreover, the Total Annual Effective Dose of exposure by oral ingestion of cassava tubers for different age groups revealed children to have the highest level of exposure with the highest mean value of 7.98 mSv. This is followed by adults and infants, which reported 5.66 and 5.38 mSv, respectively. This result is far greater than the recommended safe limits of 1 mSv (Adewoyin *et al.*, 2023). Addo *et al.*, (2013) analyzed natural radioactivity in cassava near cement factory in Volta region of Ghana, finding lower activity concentrations of radionuclide of ²²⁶Ra, ²³²Th and ⁴⁰K with an average annual effective ingestion dose of 1.23±0.09 mSvyr⁻¹. However, similar a study on natural radionuclide ingestion through foodstuffs cultivated around Sokoto Cement Company was not carried out, despite the company's operations of more than half of a century. Hassan *et al.*, (2021) assessed the level of radioactivity in staple foodstuffs as a result of contamination of soil from technological and nuclear advancement. However, the results revealed that the radionuclides in the foodstuffs were below the recommended safe limits for consumption. Therefore, this study was

targeted at analyzing and determining total annual effective dose due to radioactive exposure from food stuffs consumed by the people around Sokoto cement company, Northwestern Nigeria where cement mining activity is carried out regularly for more than half of a century. The result of this study provides a baseline data for future reference and serves as a guide in improving environmental monitoring of the study area.

MATERIALS AND METHODS

Location and Geology of the Study Area

Sokoto cement industrial area is located at Kalambaina district, 10 km south-west suburb of ancient city of Sokoto in 1960 by the then Northern Regional Government. It is the only cement production facility in the entire Northwestern zone, the most populous socio-political zone of Nigeria. It falls under Sokoto Basin which is the southern extension of the lullemeden Basin. Choosing of Kalambaina area as the location for siting the cement company was to take advantage of proximity to the required raw materials for cement production that are in abundance. The lullemeden Basin is entirely a cratonic basin created by tectonic epirogenic movements or stretching and rifting of tectonically stabilized crust during the Palaeozoic (Yelwa *et al.*, 2015). It is large synclinal structure trending NE-SE and it extends to Niger Republic, Mali, and Benin Republic (Albert *et al.*, 2016). The study area is characterized majorly with bared land and sparse vegetation. The climate is that of semi-arid with an annual average temperature of 28.3 °C (82.9 °F). The warmest months are February to April, where daytime temperatures can exceed 45 °C (113.0 °F). The rainy season is usually from June to October. From late October to February, during the 'cold season', the climate is dominated by the harmattan wind blowing Sahara dust over the land. The rock type around Sokoto Cement Company is majorly phosphate rock which is primarily associated with the Sokoto Group of sediment comprising of shale, limestone and phosphate pellets (Albert *et al.*, 2016; Obaje, 2009).

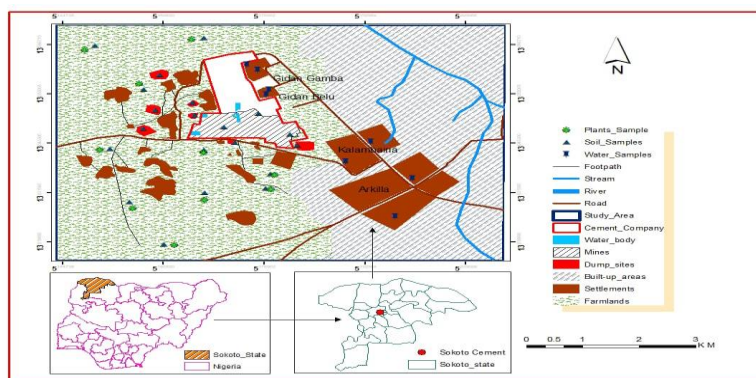


Figure 2: Map of Nigeria Showing Sokoto State and Study Area

Sample Collection, Preparation and Analysis

A total of twelve (12) samples of foodstuffs were collected directly from a group of farmlands (called Tanidawa) within the vicinity of the cement factory. Three classes of foodstuffs were collected from the study area including: Tubers of cassava (*Manihot esculenta*), and sweet potato (*Ipomoea batatas*): cereals of maize and corn (*Zea mays*); and leafy vegetables of okra (*Abelmoschus esculentus*) and pumpkin (*Cucurbita moschata*). Selection of these classes of food stuffs was because they are usually important food stuffs that contribute the most to dietary intake in Nigeria (FAO, 2021; FAOSTAT, 2000). Effort was made to ensure that at least two samples each of these were collected across the farmlands, since it was not possible to have a farmland where all the crops are grown. In each sampling point a food crop. Of mass about 1 kg was collected and put in a well labeled polythene bag. Coordinates of all the sampling points were captured using hand held GPS (Model: 78s). All samples were transported to the Center for Energy Research and Training, Ahmadu Bello University Zaria (CERT) for preparations and analysis. Each sample was spread on cardboard sheets in the laboratory. The samples were then oven dried at a temperature of about 110 °C for about half to one day. The samples were then ground into a fine powder and sieved using 2 mm mesh. The homogenized samples were filled into 500 g plastic containers (7.2 cm diameter by 6 cm height) which were hermetically sealed with the aid of PVC tape to prevent the escape of airborne ^{222}Rn and ^{220}Rn from the samples. The dimensions of the plastic containers were chosen in such a way that it suited the optimal soil mass of 350 g for analysis of bulk samples. The samples were sealed and stored for over

24 days to allow secular equilibrium to be reached between radon and its daughters. The IAEA reference materials +for gamma spectrometry (RGU-1, RGTh-1 and RGK-1) were prepared exactly as the samples.

Gamma spectrometric analysis system of NaI (TI) detector situated at low background research laboratory of Center for Energy Research and training, Ahmadu Bello University, Zaria was used for the gamma spectrometric measurements for NORMs activity concentration. The detector has a 6 cm thick lead shield, cadmium lined assembly with copper sheets for the detection of background radiation. The detector has pulse resolving duration of about 0.25 s, an incorporated preamplifier and a 1 kV external source which permits its use for high counting rates. The detector was coupled to a computer based multichannel analyzer Maestro program from ORTEC for the acquisition and analysis of the gamma spectra. The detector was calibrated with the prepared IAEA reference materials RGU-1, RGTh-1 and RGK-1 for the quantitative determination of ^{226}Ra (Bi-214 peak), ^{232}Th (TI-208) and ^{40}K (K-40) respectively in the soil samples. Each of the prepared samples was counted for 30,000 seconds in the outlined detector geometry in order to prevent the influence of background radiation from radioactive contaminants within the shielding materials of the detector assembly. The spectral energy windows used for the analysis of the NORMs were presented in Table (1).

The obtained data in counts per second were converted to conventional units of Bqkg^{-1} using calibration factors to determine the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K .

Table 1: Spectral Energy Used In the Gamma Spectrometric

Element analyzed	Gamma energy (keV)	Energy window (keV)
^{226}Ra	1764.0	1690 – 1820
^{232}Th	2614.5	2590 – 2710
^{40}K	1460.0	1380 – 1510

The equal counting time for background and sample measurement was chosen to minimize the uncertainty in the net counts (Akeozcan, 2013). The spectrum of each sample was analyzed, and the identification of unknown radionuclides was carried out by considering their peak centroid energies. The centroid energies of the peaks from the spectrum were compared with the reference gamma-ray energies (Al-Masri *et al.*, 2004).

The activity of each sample of food stuffs samples was determined by using the calibrated Na (TI) detector (Khandaker *et al.*, 2012). The activity concentration of each radionuclide in the cassava tubers sample was measured by

using the net count rates (cps) for the same counting time under the selected photo peaks, the weight of the sample, photo peak efficiency, and the gamma intensity at specific energy as given by Equation (1) (IAEA 1996).

$$A = \frac{N}{\varepsilon \times P_{\gamma} \times Tm} \quad (1)$$

Where, A is the activity of the sample in Bqkg^{-1} , N represents the net area covered by the spectrum, P_{γ} is the gamma yield, m is the mass of individual sample (kg), T is the counting time, while ε is the detector's efficiency. The lower limit of detection at 95% degree of confidence was estimated using Equation (2) (Fasanmi *et al.*, 2021).

$$LLD = \frac{N_{min}}{\varepsilon \times P_{\gamma} \times T} \quad (2)$$

Where, N_{min} stands for the minimum net area of the spectrum measured

$$N_{min} = 4.66(S_b)^{1/2} \quad (3)$$

Where S is the calculated standard error of the net count as a result of Compton scattering and effect. The Minimum Detectable Activity Concentration (MDA) was calculated using the following formular after introducing the foodstuff samples,

$$MDA = \frac{4.66(S_b)^{1/2}}{\varepsilon \times P_{\gamma} \times Tm} \quad (4)$$

Annual Effective Dose Due To Consumption of Radionuclide in Food Stuffs

Evaluation of the ingested annual effective dose estimates the radiation induced deleterious health effects associated with

consumption of radionuclides. This is because the ingested dose in the body is proportional to the total dose delivered by the radionuclides into the body. Effective dose, E ($Svyr^{-1}$) due to intake of radionuclides contaminated material was calculated using Equation (5) (Essien et al., 2021; Addo et al., 2013; DEA, 2010).

$$AED_{ing} \left(\frac{mSv}{yr} \right) = A_R IR_{ing} \sum_{j=1}^3 DCF_{ing} \quad (5)$$

Where, A_R is the mean activity concentration of radionuclides in a sample ($Bqkg^{-1}$); IR_{ing} is the soil, plant consumption rate per year. The dose conversion coefficients for the respective radionuclides are given as 2.8×10^{-4} , 6.9×10^{-4} and 6.2×10^{-6} $SvBq^{-1}$ for ^{226}Ra , ^{232}Th and ^{40}K respectively. Estimated average consumption rates for both children and adults of under study in Nigeria are presented on Table 2.

Table 2: Estimated Annual Average Consumption Rates in Nigeria ($Kgyr^{-1}$) (FAO, 2021)

food stuffs	Adults	Children
Tubers	130.2	77.5
Cereals	33.7	20.5
Leafy vegetables	22.7	11.5

RESULTS AND DISCUSSION

The mean activity concentrations of the radionuclides ^{226}Ra , ^{232}Th and ^{40}K in studied foodstuffs samples (Leafy vegetables, Cereals and Tubers) were 32.6329 ± 5.5371 $Bqkg^{-1}$, 41.7349 ± 4.7297 $Bqkg^{-1}$ and 76.2589 ± 4.9077 $Bqkg^{-1}$ whilst overall mean values were 39.9 ± 6.1347 $Bqkg^{-1}$, 57.6 ± 5.5907 $Bqkg^{-1}$ and 168.6 ± 8.4195 $Bqkg^{-1}$ respectively. These values were relatively lower than worldwide means of 67, 82 and 310 $Bqkg^{-1}$ (UNSCEAR, 2000). The values of control area were 18.53 $Bqkg^{-1}$, 33.6 $Bqkg^{-1}$ and 60.1 $Bqkg^{-1}$ for ^{226}Ra , ^{232}Th and ^{40}K respectively, which are lower than that of the study area. Comparably, the specific activities of all the studied radionuclides, were highest in tubers and lowest were in vegetables as presented on Table 3. Meanwhile, low activity concentration in cereals compared to tubers and vegetables could be attributed to the variations in the transfer factors (Essien, et al., 2021). Mean value of ^{226}Ra is not statistically significant because difference between the means (7.27 $Bqkg^{-1}$) is less than 1 combined standard deviation ($Z = 0.88$). This implies that ^{226}Ra contamination is uniformly widespread across the entire region, not just concentrated at the cement factory. For ^{232}Th it is statistically significant. This is because overall mean is significantly higher than the studied foodstuff mean by more than 2 standard deviations ($Z = 2.17$). ^{40}K is extremely statistically significant. Considering overall mean is massively higher than the studied factory samples ($Z = 9.48$), moving far past any margin of error. The results were also compared favorably with similar works, Table (4). Consequential effective dose due to consumption of radionuclides (^{226}Ra , ^{232}Th and ^{40}K) in foodstuffs for both adults and children reveals values ranged from 0.6790 $mSvyr^{-1}$ to 2.9782 $mSvyr^{-1}$ for children while the ingestion dose for adult ranged from 1.3401 $mSvyr^{-1}$ - 5.0033 $mSvyr^{-1}$, Figure (3). The values for tubers were generally higher than that of cereals and leafy vegetables. This could be attributed to the

higher transfer factors in tubers (Ocheje, & Tyovenda, 2020; Adedokun et al., 2019; Amakom et al., 2017; Addo et al., 2013). Most of these values are higher than unity (1 $mSvyr^{-1}$), a recommended limit for safety of the general public (ICRP, 2008). Even though the studied food stuffs samples represent a "cleaner" data subset compared to the world's average, they still trigger an excessive ingestion dose due to two primary factors: This could be attributed to the fact that Sokoto sedimentary basin is rich in limestone and shale (Obaje, 2009). These geological formations naturally contain trace amounts of uranium and thorium decay series, alongside potassium (Kolo, 2014). ^{226}Ra and ^{232}Th isotopes are highly toxic alpha-emitters, consuming standard regional quantities of these crops easily pushes the annual radiation dose past the public safety limit (Fasanmi, et al., 2021). ^{226}Ra shares identical chemical properties with calcium. This makes leafy vegetables and tubers actively absorb radium from the soil, mistaking it for calcium needed for cell wall development (Yachiso, et al., 2022). Internal radiation damage depends largely on the type of radiation. ^{226}Ra and ^{232}Th decay by emitting alpha particles. These particles are heavy and highly destructive to human tissue (Abba et al., 2020). Quarrying, crushing, and high-temperature clinker baking release large volumes of fine cement kiln dust (CKD) into the air. His dust settles over nearby agricultural fields (Ibrahim, 2015). Decades of deposition significantly elevate the radioactive baseline of the topsoil where local crops are cultivated (Fasanmi, et al., 2021). Also, in northern Nigeria where the study area is geographically located, cereals (like millet, sorghum, and maize) and tubers (like cassava and yam) form the absolute bulk of daily caloric intake (FAO, 2021). Hence, these could pose a concern radiological health hazard to the inhabitants of the study area being them primary consumers of these foodstuffs.

Table 3: Activity Concentration of ^{226}Ra , ^{232}Th And ^{40}K in Food Stuffs From Farmlands of the Study Area

Food Stuffs	Sample ID	Sample Type	Activity Concentration (Bqkg^{-1})		
			^{226}Ra	^{232}Th	^{40}K
Leafy vegetables	PS1	OKRO A	24.1868±3.9836	37.999±4.4432	90.7921±4.4236
	PS2	OKRO B	35.7543±5.9846	43.9525±5.1119	23.4890±1.5630
	PS3	PUMPKIN A	30.9570±5.7839	36.7203±4.4872	63.7100±3.9871
	PS4	PUMPKIN B	39.6335±6.3962	48.2679±4.8764	127.0446±9.6572
	Mean for vegetables		32.6329±5.5371	41.7349±4.7297	76.2589±4.9077
Cereals	PS5	MAIZE A	30.3622±4.8622	61.3060±6.0121	262.2942±11.9415
	PS6	MAIZE B	29.7422±4.8622	61.5266±5.9835	262.2942±13.9639
	PS7	CORN A	45.6251±6.9785	75.6396±6.6626	105.3085±6.8796
	PS8	CORN B	45.2175±6.518	73.1276±6.6448	109.3115±6.8998
	Mean for cereals		37.7368±5.8052	67.8999±6.3258	184.8021±9.9212
Tubers	PS9	CASSAVA A	54.7446±7.9926	70.9483±6.0041	354.1052±12.5689
	PS10	CASSAVA B	54.7446±7.9926	59.1761±5.6700	241.9156±10.7065
	Mean for tubers		49.3016±7.0617	63.3546±5.7166	244.7402±10.4296
Overall Mean			39.8904±6.1347	57.6631±5.5907	168.6004±8.4195
Control			18.5273±1.5425	33.6145±2.4903	60.1044±4.9741

Table 4: Mean Activity Concentrations of ^{226}Ra , ^{232}Th and ^{40}K In Compared with Other Studies

Radionuclides	Addo <i>et al.</i> , (2013)	Cardoso <i>et al.</i> , (2013)	Bello (2018)	Jibiri & Abiodun (2012)	Present study	WWA(a)
^{226}Ra (Bqkg^{-1})	15.5	64	20.4	23.9	39.9±6.13	67
^{232}Th (Bqkg^{-1})	27.9	36	37.5	18.8	57.6±5.59	82
^{40}K (Bqkg^{-1})	447	826	164	426	168.6±8.41	310

Legend: a: Worldwide Average, UNSCEAR, (2000).

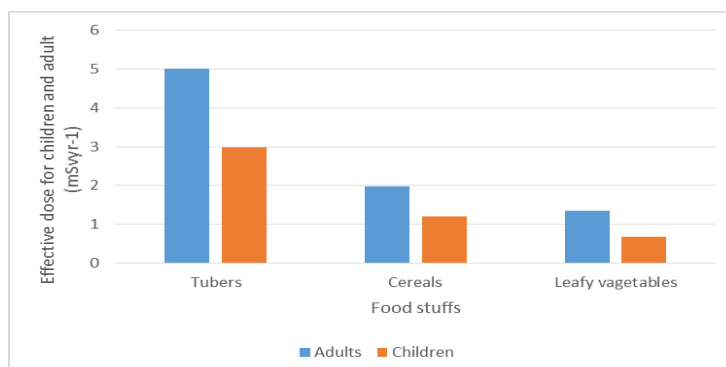


Figure 3: Effective Dose Due To Consumption of Radionuclides (^{226}Ra , ^{232}Th and ^{40}K) in Foodstuffs

CONCLUSION

This study evaluated the radiological safety of foodstuffs (leafy vegetables, cereals, and tubers) cultivated around the Sokoto cement production facility by measuring the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K . The mean activity concentrations in the immediate vicinity of the facility were found to be lower than the world's average. Statistical distinction is radiological negligible regarding public health. Despite being lower than the world average, the concentrations of highly radiotoxic alpha-emitters (^{226}Ra and ^{232}Th) in the facility's immediate vicinity remain sufficiently elevated to generate a cumulative annual committed effective ingestion dose that exceeds the internationally recommended public safety limit of unity. This may be attributed to high consumption rates of the foodstuffs, the cumulative contribution of multiple radionuclides, and the relatively high dose conversion factors associated with certain radionuclides. The findings demonstrate that beyond activity concentration, radiological risk is also influenced by dietary habits and radionuclide-specific dose coefficients. Given that these foodstuffs constitute the primary daily dietary intake for the

local population, continuous consumption of these crops poses a long-term internal radiation hazard and elevates the excess lifetime cancer risk, it is highly recommended that environmental monitoring agencies implement routine radiological assessments of the agricultural soils and water bodies in the Sokoto region. Furthermore, dust mitigation strategies at the cement facility must be optimized, and local farming communities should be educated on agricultural practices such as selective crop cultivation or soil conditioning to minimize the transfer of these specific radionuclides from the soil into the human food chain.

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