Assessment of 137Cs Soil Contamination Using Soil Samples Obtained from a Local Government in a State of Northern Nigeria

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Abstract*— Ionizing radiation is frequently released into the environment from a variety of sources which could be natural radionuclides such as Potassium isotope (40K), thorium (232Th) and its decay products (226Ra, 212Pb, etc.), and Uranium (238U) as well as artificial radionuclide such as137Cs, which eventually has a negative impact on the earth and health of the general public. The aim of this study is to determine the presence and level of 137Cs contamination in soil and computing the resulting radiation doses to the populace. With this regard, soil samples were collected to a depth of 75cm from twelve sample location in the local government area and 137Cs activity concentrations were measured using thallium-activated sodium iodide detector. The activity concentration of 137Cs was found to be in the range from 1.49±0.09Bq kg-1 to 4.46±0.26Bq kg-1 with an average value of 2.69±0.22Bq kg-1 across all twelve sample locations. The estimated external gamma-ray dose rate was calculated to be 0.21±0.02nSvh-1. The range of activity concentration of 137Cs measured was compared with other findings from around the world. The results attained for this research show that 137Cs concentration is of a lower level in some of the examine regions. The mean value calculated for external effective dose rate is seen to be below the 1.0 mSvy-1 recommended dose rate limit for the general public by the International Commission on Radiological Protection (ICRP), as well as the 0.48 mSvy-1 external gamma radiation dose per head from the natural sources of radiation evaluated by UNSCEAR. Thus, it can be said that there are no radiation risks to the people living in the area under investigation as a result of 137Cs soil pollution.

Indexed Terms*— ^{137}Caesium, Nigeria, Radiation, Radionuclides, Soil.

I. INTRODUCTION

Recent years have seen a growing body of research on measuring radioactivity in environmental samples. Ionizing radiation is frequently released into the environment from a variety of sources, which eventually has a negative impact on the health of the general public. A stream of energy known as radiation can move through space as a wave or a particle and due to component sources such as terrigenous or terrestrial, cosmogenic or extraterrestrial, and artificial or man-made radionuclides, humans are constantly exposed to environmental radiation. Natural sources including dirt, pebbles, and stones emit background radiation into the surroundings. Even water and some fruits and vegetables, such as bananas, potatoes, and almonds, may contain minute amounts of radiation. Artificially, nuclear explosions, nuclear weapon testing, and effluent discharge from nuclear facilities can all result in the release of man-made radionuclides into the environment [1]. Thus, despite the fact that radioactive radiation is an inherent component of our natural environment, it has increased to the level of a major global public concern.

The soil of our earth for instance, is an important source of nuclear radiation as numerous naturally occurring radionuclides, including the Potassium isotope (40K), thorium (232Th) and its decay products (226Ra, 212Pb, etc.), and Uranium (238U) are found in soil as inalienable components. These natural radionuclides increase radiation exposure both internally through inhalation and the food chain and externally through gamma ray emission [2]. Nuclear

technology also produces a large number of long-lived radionuclides, with 137Cs being the most prevalent one [3]. According to [4], this man-made radionuclide primarily enters the environment as a result of nuclear weapon tests, nuclear power plant accidents, and the geological repository for nuclear waste before dispersing through atmospheric convection into far-off places.

Additionally, the significant nuclear tests carried out in the late 1950s and early 1960s were responsible for the contamination from manufactured isotopes in many areas throughout the world [5]. More than 2400 nuclear weapon experiments have been carried out globally since the atomic bombing of Japan during the Second World War in 1945 [6]. However, the Chernobyl incident in 1986, which caused an estimated 3.81016 Bq of 137Cs to be discharged into the atmosphere, gave the topic of radioactive contamination global relevance and increased public awareness [7]. Long-term and short-term nuclear contamination were caused by the Chernobyl nuclear power plant accident in various regions of the world. The artificial radionuclide of concern in this work is 137Cs, the most notable artificial isotope within the class of artificial isotopes emitted as fission products as evidenced by its gamma radiation on the earth's surface. 137Cs produces gamma rays with an energy of 661.6 keV through the short-lived decay product 137Bam due to its half life of around 30.1 years.

According to [1], 137Cs's ability to emit long-lasting gamma rays, which add to external radiation exposure, is what gives it its significance. Similar to 137Cs, which may spread upon release into the stratosphere and troposphere before slowly returning to earth after a considerable amount of time, 137Cs can pollute the environment and last for decades, primarily in the soil [8], [9], [10]. Then, if it spreads to the vegetation, it can have an ecological impact. The movement of 137Cs in soils has been discussed in a number of research [11]. The type of soil, soil chemistry, organic carbon content, biological activity, climatic circumstances, and the pH-dependent absorption are only a few of the physicochemical elements that influence this process. Therefore, local terrain, precipitation, and latitude are all relevant to the deposition of radioactive fallout, including 137Cs, at any place. Due to the fact that soil particles strongly absorb and retain 137Cs, it can find its way into the diets of humans and other living things [12], [13]. The soil's qualitative and quantitative clay content, however, is the most crucial deciding element in the movement of 137Cs in soils. The mobility of 137Cs is also influenced by the components of the soil; for example, the absorption of Cs increases as the amount of organic matter in the soil decreases [14], [15]. As a result, it is well known that the soil's ability to retain 137Cs in the environment is strongly correlated with its clay concentration [16], [14].

Further, airborne deposition through atmospheric release, re-suspension of dust particles, runoff erosion by rainfall, and root uptake by plants through food chain processes are other ways that 137Cs radionuclide contamination may reach people and the environment [17], [18]. Similarly,due to its solubility, the 137Cs radionuclide can replace the Group I alkali metal elements 40K and 23Na that are involved in the ATPase pump or action potential in the body. These elements disperse evenly in the body because they have the same oxidation state but a higher affinity [19], [10], [20].

A few studies were conducted in Nigeria in various places to explore radiation measurements, including those by [21], [19], [9]. This study was carried out in Yabga East, a local government area in Nigeria's north central state of Kogi, with the objectives of confirming the presence and level of 137Cs contamination in soil and computing the resulting radiation doses to the populace to provide baseline data for hypothetical consideration. This research was also a follow-up to [21] investigation of background radiation and associated dose rates in soil samples from Yagba East Local Government in Kogi State, Nigeria.

II. MATERIALS AND METHODS

A. Instrumentation

The measurement was conducted using a thallium-activated sodium iodide detector, or NaI (TI) detector. The scintillating crystal in the detector is sodium iodide (NaI), which absorbs ionizing radiation and then emits visible or ultraviolet (UV) light. The 3-inch-diameter by 3-inch-tall cylindrical NaI crystals employed in this study are inside the detector. The light photon strikes the photocathode, which is

constructed of a substance that emits electrons in response to photons of visible light. The photomultiplying tube (PMT), which produces a signal multiplication for an electron leaving the photocathode, multiplies the electrons that are ejected from the photocathode next. The PMT's signal output is not far from the pre-amplifier, which catches the current signal and transforms it into a voltage pulse. The voltage pulse is then collected and fed into an amplifier for further amplification and pulse shaping. An output voltage pulse proportional to the charge deposited in the PMT is what the amplifier's function is to produce. The multi-channel analyzer (MCA) and Pulse Height Analysis (PHA) record and examine the output. An analog-to-digital converter, a memory with storage space, a control circuitry, and a timer are all present in the MCA channel.

B. Detector Efficiency

Since the scintillator surface's efficiency distribution is not uniform, the measurement for each sample will change depending on where it is placed on the NaI detector's surface. As a result, the NaI detector surface's efficiency profile was examined.

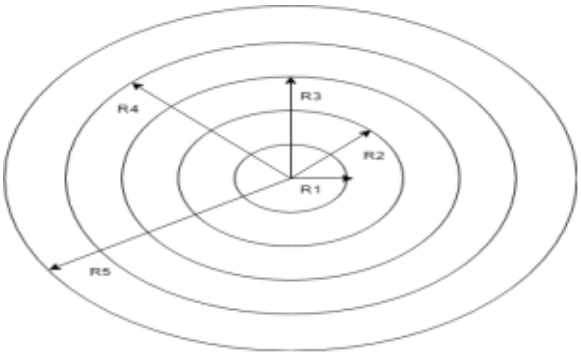

Fig. 1: Sodium iodide (NaI) detector surface containing five concentric zones

Source: [22].

The 137Cs point source was placed inside a Whirlpak® bag, and the entire container was placed on the detector surface to measure the activity (1). This was done in order to create the efficiency profile. The efficiency profile was more accurate thanks to this set-up, which replicated the measurement of an actual sample using a Whirlpak® bag. The NaI detector's surface was divided into five concentric zones (Figure 1), each with a radius of r (0.318 cm), with radii from the innermost circle to the outermost circle being r, 2r, 3r, 4r, and 5r, respectively. The Whirlpak® bag containing the 137Cs source was then placed in various zones, and the number of counts in each zone was recorded.

$$\epsilon_{detector} = \frac{N_c}{\epsilon_{geometry} \times N_p} \quad (1)$$

Where:

N_c: Number of counts per second that the detector recorded:

N_p: the amount of photons that hit the detector face

$\epsilon_{detector}$: the efficiency of the detector for a distributed sample on the detector surface

$\epsilon_{geometry}$The geometry utilized to measure a sample will determine the geometric efficiency. Here, the geometric efficiency of a point source resting flat on the detector surface is assumed to be 0.5. Thus, creating an efficiency profile for the NaI detector would help identify the detector efficiency for each sample because the container size of the soil sample could differ [22].

C. Sample Collection

Four soil samples each from three districts within a local government area of a state in north central Nigeria totaled 12 in all were collected; more information is provided in table 1. All types of dirt, both biological and non-biological, were first swept away from the surface of sampling site at each location before samples were taken at four corners and the center of a square frame about 1 m in size, and at a depth of about 75 cm, and afterwards combined to form a single sample. For easy identification, samples from each site containing soil grain weighing 0.40 to 0.50 kg were appropriately collected, labeled, and marked. After being collected, samples were put in plastic bags and brought to the lab to be measured with a NaI (TI) detector.

Table 1: Description of the sample villages and their coordinates

Sample Districts	Sample Codes	Latitude	Longitude
Igbagun	YE 01	5.71	7.84
	YE 02	5.73	7.86
	YE 03	5.74	7.88
	YE 04	5.76	7.91

Isanlu/Otedor	YE 05	5.99	8.34
	YE 06	6.00	8.37
	YE 07	6.18	8.43
	YE 08	6.26	8.49
Isanlu Makutu	YE 09	5.77	8.62
	YE 10	5.79	8.67
	YE 11	5.83	8.72
	YE 12	5.89	8.78

D. Samples Preparation and Analysis

Standard practices were used to prepare the samples. The soil samples were cleaned and allowed to dry in the sun before being sieved through a 500-mesh screen to ensure homogeneity and lower the likelihood of unfavorable attenuation during measurement. In order to completely evaporate the soil's water content, the soil samples were subsequently dried in an electric oven at a temperature of roughly 85°C for about 24 hours. To get the soil samples into a state of secular equilibrium, they were then maintained in sealed plastic containers for a month.

Furthermore, using the photo peak of the gamma-ray line with energy 661.6 keV, the activity content of 137Cs in the soil samples was calculated. Using a standard reference material obtained from the IAEA, the spectrometric system was calibrated for energy, absolute efficiency, and resolution as well as for the detection of peaks in the spectra. With a 95% level of confidence (i.e., two standard deviations), activity concentrations of 137Cs were methodically calculated and expressed relative to dry weight using the equation below [23]:

$$\text{Activity concentration} = \frac{c}{e \times i \times m} \quad (2)$$

where,

C is the net count per second .

e represents the detector's measured counting efficiency,

i is the radionuclide's gamma line's strength or intensity, and

M is the soil sample's mass, expressed in kilograms.

In order to calculate the gamma-ray dose rate due to external exposure on the basis of 137Cs activity concentration in the ground soil; it is consider that a smooth, uniform semi-infinite plane of soil contaminated with 137Cs, and a volume element of soft tissue is being exposed to a constant photon flux (F) coming from the 137Cs source concentration in the soil plane [24]. According to (3), the estimated external effective dose rate in a volume element of soft tissue at 1m height from the ground is given by the photon flux due to a single notable energy peak of 661.6keV with 85.12% abundance [24], [25], [26] as:

$$D(E) = 0.576 \times E \times \Phi(E) \times \left[\frac{\mu_a(E)}{\rho}\right]^{tissue} \quad (3)$$

For a tiny volume element of soft tissue, D(E), represents the dose rate in the soft tissue volume element in nSv h^{-1},

E: is the photon energy in (MeV)

$\Phi(E)$ is the mean photon flux at energy E in the volume element

$\left[\frac{\mu_a(E)}{\rho}\right]^{tissue}$: the energy-dependent mass absorption coefficient in cm2g-1 for the soft tissue volume element. According to [24], [25], the photon flux $\Phi(E)$ is defined based on the conservative estimate of the maximal dosage at the ground surface due to source concentration in the soil as:

$$\Phi(E) = \frac{\gamma_{FR} \times A}{2[\mu_S(E) \backslash \rho]^{soil}} \quad (4)$$

In (4) above,

$\gamma_{FR}$is fractional abundance of the gamma rays,

A is the determined (experimentally) activity concentration in Bq kg^{-1} of a radionuclide and;

$[\mu_S(E) \backslash \rho]^{soil}$ denotes the energy-dependent mass attenuation (scattering) coefficient in cm^2g^{-1} of soil that contain gamma-ray which emitt radionuclide of energy 661.6 keV.

Although soils have a wide range of chemical compositions, their relative shielding efficiency mostly depends on density and water content. As a result, the mass attenuation coefficient changes with varied soil water levels percentages and gamma-photon energy. Based on the information found in the literature, the mass attenuation coefficient for a typical soil with a density of 1.625 gcm3 and a water content of 30% by weight and 20% by volume in response to a gamma-ray energy of 661.6 keV for 137Cs was calculated to be 0.0780 cm^2g^{-1} (Jacob and Paretzke, 1986). According to the reported data (Lamarsh,1983), the mass absorption coefficient is calculated to be 0.0316cm^2g^{-1} at 661.6 keV energy for a small volume component of soft tissue used for effective dose estimation under the influence of constant photon flux

taken into account in the current study. In the 1.0 m of air above the earth layer containing the 137Cs, the build-up factor is essentially nonexistent. Also negligible is the build-up factor of photons with an energy of 661.6 keV in soft tissue that have a volume element substantially less than the mfp.

III. RESULTS AND DISCUSSIONS

Table 2 present the data of the activity concentrations of 137Cs that were determined in soil samples from twelve local government sampling locations. The activity concentrations measured vary from 1.49 Bqkg-1 at YE02 sample location to 4.46 Bqkg-1 at YE04 sample location. Whereas, the average value of 137Cs activity concentration across all 12 sample locations is 2.69 Bqkg-1. The average value of 2.69 Bqkg-1 is deviated from by 45% and 66%, respectively, by the minimum and maximum values (1.49 and 4.46 Bq kg-1) of 137Cs activity. A detailed inspection of the results from the table reveals that the concentration of 137Cs is significantly lower than the mean value of 2.69 Bq kg-1 at some sample locations, including YE01, YE02, YE04, and YE10. As opposed to the mean value of 2.69 Bq kg-1, the concentration of 137Cs is higher in various places, including YE03, YE06, YE07, YE08, YE09, YE11, and YE12. Additionally, nothing was found in the sample location of YE05, where there was no activity concentration of 137Cs.

The concentrations of 137Cs activity also differ from place to place and are not uniform, as seen in Table 2 (see Figure 2). The uneven distribution of 137Cs concentration may be a result of the variation's in topographic origins. The only anthropogenic nuclear activity close to the studied area is the use of 60Co and other transient radionuclides for industrial and medical reasons in surrounding towns. However, the nuclear fission product 137Cs has been found in several areas around the nation as a result of atmospheric fallout. In this study, the similar pattern was seen because 137Cs was discovered in all but one of the soil samples that were taken.

Table 2: 137Cs activity concentrations and associated external effective dose rates

S/N	Location	Activity concentration Bq kg^{-1}	Dose rate (nSv h^{-1})
1	YE 01	1.71 ±0.10	0.13 ±0.01
2	YE 02	1.49 ±0.09	0.12± 0.01
3	YE 03	4.22 ±0.24	0.33 ±0.02
4	YE 04	2.01 ±0.12	0.16 0.01
5	YE 05	ND	ND
6	YE 06	4.46 ±0.26	0.34 ±0.02
7	YE 07	4.02 ±0.23	0.31 ±0.02
8	YE 08	4.16 ±0.24	0.32 ±0.02
9	YE 09	3.39 ±0.20	0.26 ±0.02
10	YE 10	1.50 ±0.86	0.12 ±0.07
11	YE 11	2.80 ±0.17	0.22 ±0.01
12	YE 12	2.52 ±0.15	0.19 ±0.01
	Min.value	1.49 ±0.09	0.12± 0.01
	Max. value	4.46 ±0.26	0.34 ±0.02
	Mean value	2.69 ±0.22	0.21 ±0.02

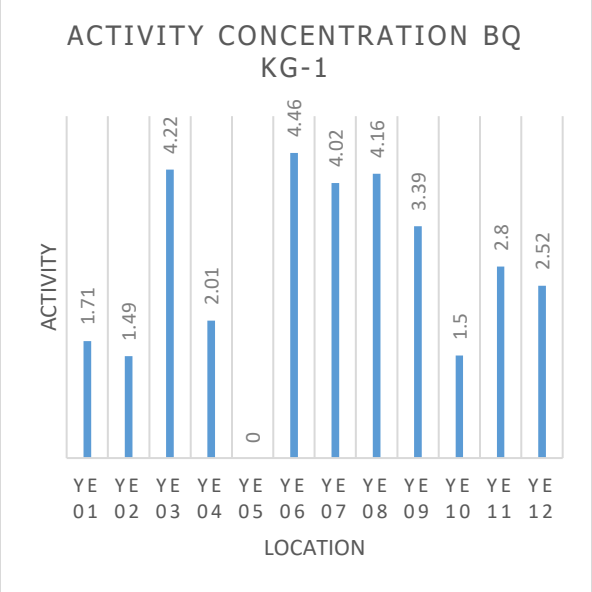

Figure 2: Activity concentration of 137Cs in the study areas

Column 4 of Table 2 displays the calculated estimates of the external effective dose rates for all twelve samples, and it can be seen that the rates range from 0.12 ±0.01 to 0.34 ±0.02 nSv h-1. From all 12 sample sites, the average estimated external dose rate is calculated to be 0.21 ±0.02nSv h-1. The International

Commission on Radiological Protection [27] suggested 1.0 mSvy-1 as the upper dose rate limit for the general public in order to reduce radiation risks. In comparison to the dose rate limit of 1.0 mSvy-1 and the average external gamma dose of 0.48 mSvy-1 received per cavity from natural radiation sources evaluated by [28], the average external gamma dose rate of 0.21 0.02nSv h-1 computed in this study is determined to be very low for any radiation risk.

Table 3 lists the 137Cs activity concentration ranges that have been recorded from several places throughout the globe. When measured values of 137Cs activity concentration in this study are compared to reported values from Majorca (Spain), Inshass-Cairo (Egypt), Algeria, Kocaeli basin (Turkey), Louisiana (USA), Montenegrin Coast (Yugoslavia), Sudan, and Northern Taiwan, it can be seen that the measured range is lower. However, as shown in a bar diagram comparison of these values with the current work in Figure 3, the values of 137Cs activity concentration measured in this study are found to be higher than the reported ranges from the Bay of Cadiz (Spain), Punjab province (Pakistan), North Western Libya, Riyadh (Saudi Arabia), and Savert (Bangladesh).

Table 3: The range of 137Cs activity concentrations in soil samples recorded around the World

Country	Concentration (Bq kg^{-1})	Reference
Majorca (Spain)	10 - 60	[29]
Inshass, Cairo (Egypt)	1.6 - 19.1	[30]
Algeria	15 - 35	[31]
Kocaeli basin (Turkey)	2 - 25	[32]
Savart (Bangladesh)	2 - 3	[32]
Louisiana (USA)	5 - 58	[32]
Bay of Cadiz (Spain)	0.5 - 5	[33]
Montenegrin coast (Yugoslavia)	1.5 - 28.4	[34]
Sudan	0 - 18.5	[35]
North-western Libya	0.9 - 1.7	[36]
Riyadh (Saudi Arabia)	0 - 2	[37]
Northern Taiwan	1.48 - 27	[38]
Punjab province (Pakistan)	1.1 - 5.3	[1]
Yagba East (Nigeria)	1.49 - 4.46	Present study

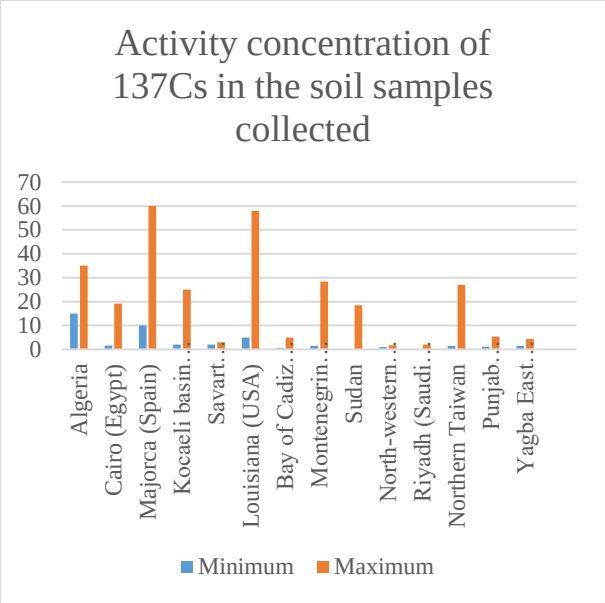

Figure 3: Activity Concentration of 137Cs in the Soil Samples around the World

CONCLUSION

As a result of atmospheric nuclear explosion tests in the 1960s, the failure of nuclear reactors like those at Chernobyl (1986, former USSR), Windscale (1957, United Kingdom), Fukushima (2011, Japan), and Kyshtym (1979, former USSR), as well as nuclear explosions like those at Kyshtym (1979), the natural environment has been contaminated by artificial isotopes. These accidents contributed to the release of large quantities of radioactive materials, particularly Caesium 137Cs, into the atmosphere and due to their unusually long half-lives, radioactive isotopes from the air have been deposited in the Earth's surface soils through precipitation and are still there. This study was carried out in a local government area of a north central Nigerian state so as to determine the presence and contamination of 137Cs in soil samples. The study found out that from twelve soil samples collected, 137Cs was found in eleven. The activity concentration of 137Cs in soil samples was found to be between 1.49±0.09Bq kg-1 to 4.46±0.26Bq kg-1, with a mean

value of 2.69±0.22Bq kg-1 across all twelve sample locations. The average annual external effective dose rate resulting from the presence of 137Cs in soil is estimated to be 0.21±0.02nSvh-1, which is low compared to the ICRP's recommended annual dose rate limit of 1.0mSv and the UNSCEAR's assessment of the annual external gamma radiation dose per person received from natural sources of radiation.

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