Radionuclide Concentration and Its Associated Radiological Hazards in the City of Ado-Ekiti Nigeria

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Abstract:

The distributions of naturally occurring radionuclides ²³⁸U, ²³²Th and ⁴⁰K at intervals of 2 cm from depth 0-10 cm in the soil of Ado-Ekiti, Nigeria were determined using well calibrated Hyper Pure Germanium Detector (HPGe). This was done in order to assess the radiological health hazards associated with the use of the soil. The mean activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K were found to be 38.2 ± 1.01 Bq kg⁻¹, 29.6 ± 0.9 Bq kg⁻¹ and 1018.3 ± 23.6 Bq kg⁻¹, respectively. Radium equivalent (Raeq), External Hazard Index (Hex), Internal Hazard Index (Hin), Absorbed Dose Rate (D) and Annual Effective Dose Rate (AEDR) were found to be, 159.1 Bq kg⁻¹, 0.43 Bq kg⁻¹, 0.53 Bq kg⁻¹, 85.5 nGy h⁻¹ and 0.42 mSv y⁻¹ respectively on the average. The results of the radiological indices and dose rates obtained in this study were all higher than their worldwide mean values but lower than their maximum recommended limits indicating that the use of soil for farming purposes and as building materials, and molding of earthen wares do not constitute any excessive radiological hazards. Therefore, the results of this study could serve as baseline data upon which future environmental radiometric monitoring initiatives could be based.

Keyword: Radionuclide, radiological hazard, radiological indices, soil, absorbed dose

Introduction

The material from which the earth was formed, about many billion years ago contains many unstable nuclides (Scholten and Timmermans; Raad and Hayder, 2011). Consequently, human beings are continually being exposed to ionizing radiation from natural sources in a way they can hardly avoid. In most cases, the exposure of every individual from this natural source exceeds that from all man-made sources put together (UNSCEAR, 2008). Hence, natural radioactivity from naturally occurring radioactive materials NORMs is widely spread in the earth's environment and it exists in various geological formations such as soils, rocks, water, sediment, air and in building materials. Artificial radionuclides from nuclear weapon tests, nuclear accidents, medical and industrial applications also contribute about 23% of human exposure. Exposure from nature comes from the naturally occurring radioactive isotopes of ²³⁸U, ²³²Th and their progenies as well as ⁴⁰K (Chikasawa et al., 2001; Tzortzis et al. 2004; UNSCEAR, 2000 ; Shetty and Narayana, 2010; UNSCEAR, 1993). The contribution of radiation from soil to human exposure can either be whole body due to external radiation originating directly from primordial radionuclides present in soil or internal due to inhalation (Jibiri and Okeyode, 2012; Ngachin et al., 2007). The internal exposure to radiation, affecting the respiratory track, is due mainly to radon and its decay products which emanate from primordial radionuclides in soil, water and food items (Hameed et al., 2014; Tso and Leung, 2000, UNSCEAR, 1993). Long-term exposures

to radioactivity and inhalation of radionuclides have serious health effects such as chronic lung cancer and leukemia (Qureshi et al., 2014, Masita et al., 2008). Estimation of the radiation dose distribution is therefore important in assessing the health risk to a population and this will also serve as a reference in documenting changes to environmental radioactivity in soil due to anthropogenic activities (Obed et al., 2005).

Method

Soil samples were collected from randomly selected points in Ado-Ekiti. At each of the points, samples were collected between depths 0-2, 2-4, 4-6, 6-8, 8-10 cm in the soil after the surface had been cleared of decaying organic matter (IAEA, 1991). The samples were sun dried until they maintained a constant weight. The samples were then ground using a mole grinding machine and sieved with 2mm wire mesh to obtain a fine texture of the samples. The sieved soil samples were weighed 250g lot into a cellophane bag (NRC, 1990). The packaging was done with the use of thick cellophane bag with a press-and-key mouth to make it airtight. The cellophane bags were properly labeled to indicate the point and depth where the samples were collected.

The sieved soil samples were sealed in 76cm³ container and subjected to gamma ray spectroscopy to determine the activity. Gamma spectroscopy measurements were carried out with coaxial-type Germanium, detectors (HPGe) by Canberra Industries Inc. The detector has a 50% relative efficiency and resolution of 2.4 keV at 1.33 MeV. The detector was properly shielded in lead castle. Calibrations of the measuring systems had been carried out with certified reference standards for various radionuclides. Spectral analyses were performed with the Genie 2K spectrometry software, version 2.1 (Canberra Industries Inc.)

Each sample was counted for 24 hours to achieve minimum counting error. Specific activity of each radionuclide in soil samples was expressed in Bq kg⁻¹ of dry mass of soil and corrected for the time elapsed since the samples were collected in the field (IAEA, 1991). The average of the values obtained for similar depths were taken to be the reading of that depth in the city of Ado-Ekiti.

Discussion

Radium equivalent: Bq Kg⁻¹

To represent the activity levels of ²³⁸U, ²³²Th and ⁴⁰K and take into account the radiological hazards associated with them, a common radiological index has been introduced. This index is called radium equivalent activity (Raeq) and is mathematically defined by UNSCEAR 2000.

Raeq = Au + 1.43ATh + 0.0077Ak

(1)

Where A_U , A_{Th} and A_K represent the activity concentrations of Uranium, Thorium and Potassium respectively. The permissible limit of Raeq is 370Bq.kg⁻¹ in soil that contain ²³⁸U, ²³²Th and ⁴⁰K measured in Bq.kg⁻¹ (UNSCEAR 2000)

Hazard Index: Bq/kg

A widely used hazard index (reflecting the external exposure) called the external hazard index, Hex is defined as follows (UNSCEAR, 2000)

 $Hex = \frac{Au}{370} + \frac{ATh}{259} + \frac{Ak}{4810}$

(2)

In addition to external hazard index, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified

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by the internal hazard index, Hin, which is given by the equation (Raad et. al., 2011; Isinkaye et. al. 2013)

 $Hin = \frac{Au}{185} + \frac{ATh}{259} + \frac{Ak}{4810}$ (3) Where A_U, A_{Th} and A_K are the specific activities of ²³⁸U, ²³²Th and ⁴⁰K, respectively. The

where A_U , A_{Th} and A_K are the specific activities of $U^{-1}U$, $U^{-1}h$ and $U^{-1}K$, respectively. The values of the indices (Hex, Hin) must be less than unity for the radiation hazard to be negligible

Absorbed Dose

In estimating the heath risk associated with exposure to radiation from radioactivity in the soil, it is necessary to convert the activity concentration to absorbed gamma dose rates in air at 1m above the ground surface. This is calculated using the equation given by Beck et al. 1972 as shown in the following expression;

(4)

 $D = 0.042S_{K} + 0.429S_{U} + 0.666S_{Th}$

where *D* is the absorbed dose rate in air (nGy h⁻¹), and S_K , S_U , S_{Th} are the soil specific activity concentration of ⁴⁰K, ²³⁸U and ²³²Th respectively taking ²¹⁴Bi and ²⁰⁸Tl as indicators for ²³⁸U and ²³²Th respectively.

Annual Effective Dose Rate

In order to make an estimate of the annual effective dose outdoor, there is need to take into account the conversion coefficient from absorbed dose in air to effective dose and the outdoor occupancy factor. In the UNSCEAR recent reports (1993, 2000), the committee used 0.7 Sv Gy⁻¹ for the conversion coefficient from absorbed dose received by adults, 0.2 for the outdoor occupancy factor (this suggests that an average person stays for about 5hours outside daily). Thus the effective dose rate outdoor in units of Sv per year is calculated from the formula (UNSCEAR 2000):

$$AEDR = D(\gamma) \times N(h) \times O_f \times C_f$$
(5)

Where *AEDR* is annual outdoor effective dose rate (μ Sv y⁻¹), $D(\gamma)$ is the absorbed dose rate in air (nGy h⁻¹), N(h) is number of hours in a year (24x365.25), O_f is the outdoor occupancy factor, and C_f is the conversion factor (Sv Gy⁻¹).

The values and the associated error in the concentration of radionuclides measured at intervals of 2 cm to the depth of 10cm in the soil of Ado-Ekiti Nigeria are presented in Table 1. The corresponding contributions of each depth to radiological hazard indices are shown in Table 2. Figure1 shows the graphical representation of radionuclide concentration across the soil depth. The mean activity concentrations of the radionuclides in the samples are also given in Table 1. The gamma lines identified with reliable regularity belonged to the decay series of naturally occurring radionuclides headed by ²³⁸U and ²³²Th. Other gamma lines belong to ²³⁵U and the singly occurring natural radionuclide ⁴⁰K as well as the artificial radionuclide ¹³⁷Cs which was detected in very small concentration. Its values ranged from 0.09 to 0.80 Bq kg⁻¹. The average value concentration detected for ⁴⁰K is the highest (1018.3± 23.6 Bq kg⁻¹) followed by ²³⁸U (38.2± 1.01 Bq kg⁻¹) and the least was ²³²Th (29.6±0.90 Bq kg⁻¹) The highest value obtained for ⁴⁰K might be due to the fact that the use of NPK fertilizer for agricultural purpose is predominant in the environment. This has considerably added to the soil potassium (Essiet et. al.). The permissible limit of Raeq is 370Bq.kg⁻¹ in soil that contain ²³⁸U, ²³²Th and ⁴⁰K measured in Bq.kg⁻¹ (UNSCEAR 2000). The mean Raeq for the soil in this study is 159.1 which fall below the permissible limit of 370Bq.kg⁻¹ given by UNSCEAR, 2000. The results also shown by the

values of Hex and Hin indicate that the radiation hazard contributed by ${}^{40}K$, ${}^{238}U$ and ${}^{232}Th$ is negligible because the average values are less than unity. 0.43 and 0.53 Bq kg⁻¹ for Hex and Hin respectively.

The presence of man-made radionuclide ¹³⁷ Cs could be the result of the Chernobyl nuclear accident of 1986 and the weapon tests in the Sahara region. The detection of this in the study location could be due to the proximity of the study location to the location of the incident. It is therefore crucial to carry out regular monitoring exercise on the seasonal variation of the radioactivity concentration of caesium in Ado-Ekiti. This is because caesium is highly carcinogenic.

Conclusion

The results of the radiological indices and dose rates obtained in this study were all higher than their worldwide mean values but lower than their maximum recommended limits. This indicates that the use of soil for farming purposes, molding of earthen wares and construction of building do not constitute any excessive radiological hazards to the people in the community.

Table 1: Radionuclide Concentration (Bq kg⁻¹) across the soil at intervals of 2cm to the depth of 10cm

| Soil | 40 K | ²⁰⁸ Tl | ²¹⁰ Pb | ²¹² Bi | ²¹² Pb | ²¹⁴ Bi | ²¹⁴ Pb | 224 Ra | ²²⁶ Ra | ²²⁸ Ac | 228Th | 234Th | ²³⁵ U | ¹³⁷ Cs |
|-------|------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------|-------------------|-------------------|-------|-------|------------------|-------------------|
| Depth | | | | | | | | | | | | | | |
| Depui | | | | | | | | | | | | | | |
| (cm) | | | | | | | | | | | | | | |
| | | | | | | | | | | | | | | |
| 0-2 | 1065.7± | 30.2 | 70.3 | 104.8 | 101.8 | 33.9 | 37.6 | 89.8 | 19.4 | 93.9 | 171.6 | 74.2 | 4.4 | 0.09 |
| | 25.8 | +0.9 | +5.0 | + 6.6 | +3.1 | +0.95 | +1.3 | + 7.6 | +49.8 | +2.8 | +11.4 | +3.4 | +3.0 | +0.2 |
| | 2010 | _0.0 | = 0.10 | _ 010 | _ 0.1 | _0.70 | = 110 | _ / 10 | = | = =.0 | | | _0.0 | _ 0 |
| | | | | | | | | | | | | | | |
| 2-4 | 900.3 | 24.9 | 18.9 | 97.1 | 87.1 | 35.5 | 34.8 | BDL | 79.6 | 69.4 | 224.5 | 44.9 | 4.9 | 0.21 |
| | + 21 2 | +0.8 | +44 | + 5 6 | + 2 2 | +1.0 | +1.0 | | +65 | +2.0 | +12.1 | +24 | +0.4 | +0.2 |
| | ± 21.2 | -0.0 | | ± 5.0 | - 2.2 | ± 1.0 | ± 1.0 | | ± 0.5 | ± 2.0 | ±12.1 | - 2.4 | -0.4 | ± 0.2 |
| 1.6 | 1020.0 | 20.7 | 16.2 | 105.0 | 90.2 | 41.5 | 44.4 | DDI | 105.2 | 05 1 | DDI | 76.2 | (5 | 0.20 |
| 4-0 | 1030.9 | 30.7 | 10.2 | 105.9 | 89.5 | 41.5 | 44.4 | BDL | 105.2 | 85.4 | BDL | /6.2 | 0.5 | 0.20 |
| | ± 24.9 | ±0.9 | ±10.5 | ± 6.4 | ± 4.5 | ± 1.1 | ± 2.4 | | ± 8.4 | ± 3.6 | | ± 6.5 | ±0.5 | ± 0.2 |
| | | | | | | | | | | | | | | |
| 6-8 | 1055.0 | 31.3 | 49.7 | 115.8 | 101.2 | 41.1 | 42.8 | 84.7 | 93.8 | 88.2 | 143.9 | 66.8 | 5.8 | 0.80 |
| | +21.6 | +0.8 | + 5 3 | +6.8 | +2.8 | +1.1 | +17 | +7.6 | +64 | +2.9 | +10.9 | +39 | +0.4 | +0.2 |
| | = 21.0 | _0.0 | _ 5.5 | = 0.0 | = 2.0 | | = 1.7 | = 7.0 | = 0.1 | = 2.9 | _10.9 | _ 5.9 | _0.1 | _ 0.2 |
| 8-10 | 1039.3 | 31.1 | 12.6 | 101.6 | 111.9 | 39.6 | 44.2 | BDL | 95.1 | 83.7 | 145.1 | 91.9 | 5.8 | 0.50 |
| | + 24.6 | +0.9 | +95 | +69 | +2.8 | +12 | +13 | | +71 | + 2 7 | +18.1 | +4.7 | +0.4 | +0.3 |
| | ± 27.0 | ±0.7 | ÷ 7.5 | ± 0.7 | ±2.0 | ÷ 1.2 | ± 1.5 | | ± /.1 | ± 2.1 | ±10.1 | ····/ | ±0.4 | ± 0.5 |
| Mean | 1018 3 | 29.6 | 33.5 | 105.0 | 98.3 | 38.2 | 40.8 | 34.9 | 78.6 | 53.2 | 137.0 | 70.8 | 55 | 0.40 |
| wican | 1010.5 | 20.0 | 55.5 | 105.0 | 2.1 | 1.01 | -10.0 | 57.7 | 15.0 | 55.2 | 137.0 | 10.0 | | .0.0 |
| | ± 23.6 | ±0.9 | ± 6.9 | ± 0.5 | ± 5.1 | ± 1.01 | ± 1.5 | ± /.6 | ±13.6 | ± 2.8 | ± 1.2 | ± 4.2 | ±0.9 | ±0.2 |
| | ± 23.6 | ±0.9 | ± 6.9 | ± 6.5 | ± 3.1 | ±1.01 | ± 1.5 | ± 7.6 | ±15.6 | ± 2.8 | ± 7.2 | ± 4.2 | ±0.9 | ±0.2 |

BDL (Below Detectable Limit)

Table 2: Contribution of each depth to radiological hazard Indices

| Depth (cm) | Radium | Hex | Hin | D | Outdoor | |
|------------|----------------------------------|----------------|----------------|----------------|-------------------|--|
| | Equivalent | $(Bq kg^{-1})$ | $(Bq kg^{-1})$ | $(nGy h^{-1})$ | AEDR | |
| | $(\mathrm{Bq} \mathrm{kg}^{-1})$ | | | | $(\mu Sv y^{-1})$ | |
| 0-2 | 159.2 | 0.43 | 0.52 | 85.0 | 0.42 | |
| 2-4 | 140.4 | 0.39 | 0.48 | 75.8 | 0.37 | |
| 4-6 | 164.8 | 0.44 | 0.55 | 88.7 | 0.44 | |
| 6-8 | 167.1 | 0.45 | 0.56 | 89.8 | 0.44 | |
| 8-10 | 164.2 | 0.45 | 0.55 | 88.1 | 0.43 | |
| Mean | 159.1 | 0,43 | 0.53 | 85.5 | 0.42 | |



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