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RESEARCH ARTICLE

MEASUREMENTS OF LEVELS OF NATURAL RADIONUCLIDES IN THE SOIL IN ZUWAYE, AND SHASHAMENE AREA, ETHIOPIA, USING GAMMA RAY SPECTROMETRY

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ABSTRACT

The activity concentration of Naturally Occurring Radio-nuclides such as ^{238}U , ^{232}Th and ^{40}K in the collected soil sample from the selected area were measured by using Gamma ray spectroscopy employed by HPGe detector to perform the measurements. The average activity concentration values was found to be 46.93, 110.6, 847.65 and 2.43Bq/Kg for ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs respectively. These values were higher than the world average values given by UNSCEAR (2000). The average associated radiological hazard parameters indices Raeq, Hex, Hin, Dout, Din, AEDE and $I\gamma$ of natural radio-nuclides were determined to be as follow 248.27, 0.728, 0.86, 0.185, 1.035, 1.22 and 1.98 Bq/Kg respectively. Some results such as Dout, Din, AEDE and $I\gamma$ were found to be higher than the recommended world average values UNSCEAR (2000) and others were Raeq, Hex and Hin below the standard. Accordingly, the investigated soil sites were not radiation hazard free.

INTRODUCTION

Natural radioactivity is a source of continuous exposure to human beings. It is present in the human environment due to the presence of cosmogenic and primordial radionuclides in the Earth's crust. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in the soil of each region in the world [1]. Cosmogenic radionuclides are produced by the interaction of cosmic-rays with atomic nuclei in the atmosphere, while primordial ones (terrestrial background radiation) were formed by the process of nucleo-synthesis [2]. The great interest expressed worldwide for the study of naturally occurring radiation and environmental radio activity has led to interest in extensive surveys in many countries. Natural sources still contribute almost 80% of the collective radiation exposure of the world's population. There are many sources of radiation and radioactivity in the environment. Gamma radiation emitted from naturally occurring radionuclides, also called terrestrial background radiation, represent the main external source of irradiation of the human body [3,4]. Significant amount of man-made radionuclides ^{137}Cs and ^{90}Sr may be also present in the soil and plant as a result of testing of nuclear weapons in the atmosphere, accidents, such as Chernobyl accident, and the routine discharge of radionuclides from nuclear installations. The contribution of other nuclides to the total activity is negligible [4]. Once present in the environment, these radionuclides, whether natural or artificial, are available for

uptake by plants and animals and so make their way into the food chain [5]. Human beings are exposed to radiation from sources outside their bodies; mainly, cosmic rays and gamma ray emitters in soils, building materials, water, food, and air. Studying the levels of radionuclide distribution in the environment provides essential radiological information [3]. The amount of radioactivity in soil varies widely; hence it is important to monitor the terrestrial background radiation mainly due to natural radionuclides in soil [6]. Soil from waste dump sites may contain naturally occurring radionuclides in significant amounts and the resulting external radiation exposure pathway to the population has been subjects for study [7]. Many studies worldwide have measured the activity concentration of natural radionuclides in soil to ascertain the levels of contamination [3, 5-23]. The present work aims to estimate the activity concentration of radionuclides ^{226}Ra , ^{232}Th , and ^{40}K in soil samples collected from Shashamene and Zuwaye Ethiopia, and to evaluate the radiological indices and their effects on the population who live in this environment. Therefore, the results were used to assess the potential radiological hazards associated with these soils by estimating the radiological indices.

METHODS AND METHODS

Sample collection and sample processing: A total of 3 surface soil samples in 4 major sites were collected. The top layers of the soil which contained wastes that are yet to decompose were removed. Soil samples were collected to a

depth of 5 cm using a coring tool that was thoroughly cleaned and dried before each sample was collected. Ultimate care was taken in the extraction of soil sections to avoid mixing or cross contamination of soil samples. About 2 kg of each sample were collected in a plastic bag at the sampling points. The soil samples were processed according to the procedure recommended by the IAEA. Soil samples were well mixed after removing exotic materials such as pieces of stones and gravel. The samples were weighed and then dried in an oven at 110°C. After shaking thoroughly, the samples were sieved with a 1 mm mesh screen [6]. The samples were dried, sieved, packed in 1 L Marinelli beaker and sealed for 4 weeks to reach secular equilibrium between ^{226}Ra (daughter of ^{238}U) and ^{232}Th with their daughter nuclei. This means in order to allow for radon and its short-lived progenies to reach secular radioactive equilibrium prior to gamma spectroscopy [13].

Theoretical Calculations

The activity concentration: The activity concentrations of the radionuclides in the measured samples were computed using the following relation [13]:

$$C = \frac{C_a}{I \times \varepsilon_{ff} \times M_s}$$

Where C_a is the net gamma counting rate (counts per second), ε_{ff} the detector efficiency of the specific γ -ray, I is the intensity of the γ -line in a radionuclide and M_s is the mass of the sample (kg).

Radiological Effects

The Radium Equivalent Activity (Raeq): For the purpose of comparing the radiological effect or activity of materials that contain ^{226}Ra , ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them, a common index termed the radium equivalent activity (*Raeq*) is used. This activity index provides a useful guideline in regulating the safety standards on radiation protection for the general public residing in the area under investigation. The *Raeq* index represents a weighted sum of activities of the above mentioned natural radionuclides and is based on the estimation that 1 Bq/kg of ^{226}Ra , 0.7 Bq/kg of ^{232}Th , and 13 Bq/kg of ^{40}K produces the same gamma radiation dose rates. The index is given as:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.007C_K$$

Where C_{Ra} , C_{Th} and C_K are the average activity concentration in the sample in Bq/kg of ^{226}Ra , ^{232}Th , and ^{40}K respectively [13].

Absorbed dose rate in air: The effects of gamma radiation originating from radioactive sources in the environment are generally expressed in terms of the total gamma radiation absorbed dose rate in air, D_r . The values of D_r in air and 1 m above the ground level are calculated from the measured activity concentrations of ^{238}U , ^{232}Th , and ^{40}K radionuclides using the following semi empirical formula [25, 26].

$$D_r(nGy h^{-1}) = 0.427xAU + 0.662xATH + 0.043AK$$

Eq. (3) was modified to include the contributions of artificial radionuclides of cesium, (^{137}Cs), as well as cosmic radiation via the following equation [25].

$$D_r(nGy h^{-1}) = 0.427xAu + 0.662xATH + 0.043xAK + 0.03XACs + 34$$

Here 0.427, 0.662, and 0.043 are the dose rate conversion factors to convert the activity concentrations of ^{238}U , ^{232}Th , and ^{40}K and ^{137}Cs radionuclides into absorbed dose rates as proposed by UNSCEAR [27]. Basically, these factors are representative of the absorbed dose rates in air per unit activity per unit of soil mass, in units of nGy h⁻¹ per Bq kg⁻¹.

Annual effective dose equivalent: The annual effective dose equivalent (*AEDE*) received by individuals was calculated from the calculated values of D_r by applying the dose rate conversion factor of 0.7 Sv Gy⁻¹ and the occupancy factors of 0.2 (5/24) and 0.8 (19/24) for outdoors and indoors, respectively [27]. The annual effective outdoor doses, D_{out} ; the annual effective indoor doses, D_{in} ; and total annual effective doses, D_{tot} , were calculated according to the following equations [28].

$$D_{out}(mSv y^{-1}) = D_r(mGy h^{-1}) \times 24 \times 365.25 \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6}$$

$$D_{in}(mSv y^{-1}) = D_r(mGy h^{-1}) \times 24 \times 365.25 \times 1.4 \times 0.8 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6}$$

$$D_{tot}(mSv y^{-1}) = D_{out} + D_{in}$$

The external and internal hazard index: The external (H_{ex}) and internal (H_{in}) hazard index due to the emitted γ -rays of the soil samples were calculated and examined according to the following criterion:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1$$

$$\text{And } H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810}$$

The value of H_{ex} must be lower than unity in order to keep the radiation hazard insignificant. This is the radiation exposure due to the radioactivity from a construction material, limited to 1.5 mGy.y⁻¹. The maximum values of H_{ex} equal to unity correspond to the upper limit of *Raeq* (370 Bq.kg⁻¹) [20].

Radioactivity level index: The radioactivity level index, I_γ , is generally used to assess the hazardous level of radionuclides in the human body when exposed to an amount of external (indoor or outdoor) annual effective doses of γ -radiations decayed from radioactive nuclide in the soil. This index is very important for quality control of γ -radiation annual effective dose and in monitoring radiation inside human body, to ensure that such radiation does not exceed the worldwide permissible high dose values [29]. Values of I_γ can be calculated according to the semi empirical formula [30,31].

$$I_\gamma = \frac{C_{Ra}}{150 \text{ Bq/Kg}} + \frac{C_{Th}}{100 \text{ Bq/Kg}} + \frac{C_K}{1500 \text{ Bq/Kg}}$$

The assessed values of I_γ must be less than or equal to 1 to make sure the soil environment is generally safe or hazard free.

EXPERIMENTAL RESULTS AND DISCUSSION

Detector Characterization

Energy Calibration: The detector calibration was performed using a certified standard reference mixed material ¹³⁹Ce (166 keV), ¹¹³Sn (392 keV), ⁸⁵Sr (514 keV), ¹³⁷Cs (662 keV), ⁸⁸Y (898 and 1836 keV) and ⁶⁰Co (1173 and 1332 keV) in the energy range (166 - 1850) keV [13]. The channel number and associated peaks were recorded. The graph between channel number and the energy was plotted. From the energy calibration figure the relation between channel and energy was linear.

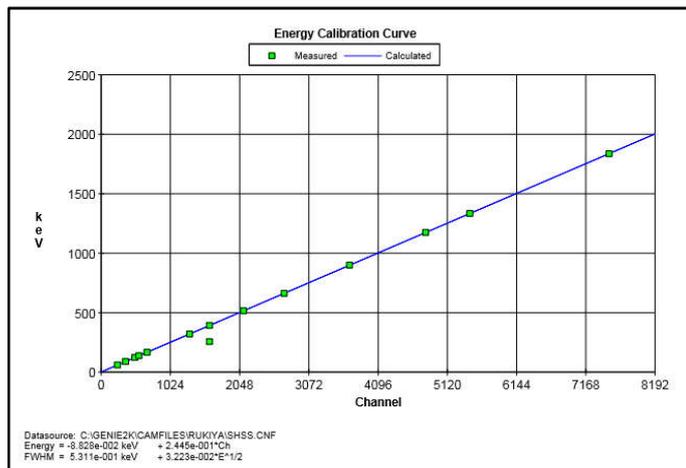


Figure 1. Energy calibration relation between channel numbers and corresponds to energy

Energy versus efficiency curve: Efficiency Calibration is the ratio between the number of counts in the full energy peak and number of radiation incident on the detector [35]. Two types of efficiency can be determined

- a. Absolute Photo Peak efficiency
- b. Intrinsic Photo Peak Efficiency

In this study the relative efficiency was considered. The Energy efficiency calibration curve beyond 1850 keV was constructed using different energy peaks of ²²⁶Ra in order to cover the range from 60 up to 2000 keV [13].

The graph between efficiency and the energy was plotted.

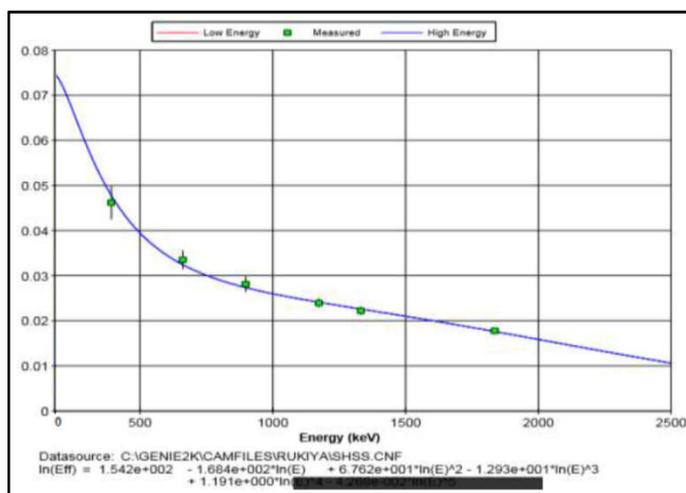


Figure 2. Detector energy Vs efficiency curve

The activity concentration: The results of analysis of activity concentration of ²³⁸U, ²³²Th, ⁴⁰K and ¹³⁷Cs radionuclides in soil samples for different locations of the study area are presented in (Table 1). The activity concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs in the samples were determined by standard gamma spectrometry using aHPGe detector (Ortec) with a 70% relative efficiency and a resolution 1.9 keV for the 1332.5 keV ⁶⁰Co gamma line and MCA with 2000 channel. The standard source packed in the Marinelli beaker had the same geometry as that used for measured samples. The background radiation and the samples were counted 36,000s. The 295.21 and 351.92 keV of ²¹⁴Pb and 609.31, 1120.29 and 1764.49 keV of ²¹⁴Bi gamma ray lines were used to determine the ²³⁸U activity concentration. The ²³²Th activity concentration was determined using 238.63 keV of ²¹²Pb, 911.21 and 968.97 keV of ²²⁸Ac gamma lines. The activities of ⁴⁰K and ¹³⁷Cs were determined directly from the 1460.8 and 661.6 keV gamma lines, respectively. The net count rate under the most prominent photo peaks of all radio- nuclides daughter peaks were calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. Then the activity of the radionuclide is calculated from the background subtracted area prominent gamma ray energies [17].

Table 1. The measured Activity concentration level of natural radionuclides in the soil samples collected from different sites located in Shashemene and Zuwaye

Sample ID	Activity Concentration		
	²³⁸ U	²³² Th	⁴⁰ K
SHSS	54.02126 ± 3.423728	151.2349 ± 11.76711	784.584 ± 32.6551
BDSH	27.60476 ± 1.761418	80.88988 ± 6.657935	806.968 ± 39.7541
BDH	37.6856 ± 81.08334	76.78563 ± 165.2413	858.251 ± 36.05
SHKS	56.92794 ± 3.666244	117.0734 ± 9.374495	979.578 ± 40.8789

As we can be seen from Table 1, the value of measured activity concentration of ²³⁸U, ²³²Th, ⁴⁰K and ¹³⁷Cs varies from 27.60 to 58.4, 76 to 151.23, 784.58 to 979.57 and 1.13 to 3.17Bq/Kg respectively. The average activity concentration of ²³⁸U, ²³²Th and ⁴⁰K was 46.93, 110.6, 847.65Bq/Kg, respectively. These values were higher than the recommended world average values for ²³⁸U, ²³²Th, ⁴⁰K, respectively which are 35, 30 and 400 Bq/Kg for ²³⁸U, ²³²Th, ⁴⁰K respectively [1, 4]. It is also observed that the measured activity concentration of ⁴⁰K exceeds markedly the values of both Uranium and Thorium, as it is the most abundant radioactive element under consideration. Moreover the excessive use of the Potassium containing fertilizers in the area adjacent to the sampling sites may contribute to the higher values of ⁴⁰K activity. The activity concentrations of the artificial radionuclide ¹³⁷Cs were measured for all collected soil samples in order to assess the amount of fallout radionuclide in such locations; they are given in Table 1. The obtained activity concentration values of ¹³⁷Cs in all collected soil samples were found to range from 1.13 Bq kg⁻¹ to 3.172 Bq kg⁻¹ with an average value of 2.43Bq kg⁻¹. The minimum activity concentration value of ¹³⁷Cs was obtained for a soil sample collected from zuwaye, whereas the maximum value was measured in a soil sample collected from shashemane area. Thus, the impact of the artificial radionuclide and the corresponding additional external radiation exposure to the population were almost negligible. Consequently, the measured activities of ¹³⁷Cs confirmed no hazard effects due to

^{137}Cs radionuclides to the people living around the sites where soil samples were collected.

Radiological Effects: The γ -radiation hazards associated with these samples were assessed according to different indices. The calculated data for R_{aeq} , D_r , D_{out} , D_{in} , H_{ex} and I_γ of the collected soil samples are shown in table 2.

Radium equivalent activity

It's calculated through the following relation:

$$R_{aeq} = C_{Ra} + 1.43C_{Th} + 0.07C_k$$

Where C_{Ra} , C_{Th} and C_k are the activities concentration of Ra-226, Th-232 and K-40 in Bq/kg, respectively. In calculating R_{aeq} values, the average activity concentrations of 370 Bq kg $^{-1}$, 259 Bq kg $^{-1}$, and 4810 Bq kg $^{-1}$ used for ^{238}U , ^{232}Th , and ^{40}K radionuclides, respectively, were assumed to produce the same gamma dose rate [32].

Table 2. The associated radiological hazard parameters indices R_{aeq} , H_{ex} , H_{in} , D_r and I_γ of natural radionuclides.

Sample ID	R_{aeq} Bq/Kg	D_r (nGy/h)	D_{out} (mSv/y)	D_{in} (mSv/y)	AEDE	H_{ex}	H_{in}	I_γ
SHSS	330.7	191.1	0.234	1.312	1.546	0.86	1.034	2.395
BDSH	205.4	100.1	0.123	0.69	0.813	0.55	0.63	1.531
BDH	155.8	137.9	0.169	0.95	1.119	0.59	0.68	1.591
SHKS	299.8	143.9	0.177	0.989	1.166	0.81	0.963	2.203
KSS	249.7	179.8	0.2206	1.235	1.46	0.83	0.985	2.228

The values of radium equivalent for different soil samples in area under investigated were calculated by using equation above these values presented in table 2 and values ranged from 155.8 to 330.7 Bq/kg with average value 248.27 Bq/kg which is lower than the recommended maximum value 370 Bq/kg [13].

Absorbed dose rate in air: The calculated values of D_r were found to vary from 100.036 to 191.021 nGy h $^{-1}$, with an average value of 150.53 nGy h $^{-1}$. The measured average absorbed dose rate in the air and the measured average value of the representative level index are higher than the recommended international levels of 55 nGy h $^{-1}$ [1, 4]. Such locations are not suitable for human residency, and constructed homes should be removed.

Annual effective dose equivalent: The calculated indoor, outdoor, and total $AEDE$ values are presented in Table 2. The calculated values for D_{out} , D_{in} , and D_{tot} averages were respectively 0.185, 1.035, and 1.22 mSv year $^{-1}$. In comparison to global measured values, these values were all higher than the assigned worldwide values of 0.08, 0.42, and 0.50 mSv year $^{-1}$, respectively [1]. The locations from which the soil samples collected were all not safe according to the Radiation Protection, and such locations cannot be classified as hazard free [34].

The external and internal hazard index: The external (H_{ex}) and internal (H_{in}) hazard index due to the emitted γ -rays of the soil samples were calculated and examined according to the following criterion:

The value of H_{ex} must be lower than unity in order to keep the radiation hazard insignificant. This is the radiation exposure due to the radioactivity from a construction material, limited to 1.5 mGy \cdot y $^{-1}$. The maximum values of H_{ex} equal to unity correspond to the upper limit of R_{aeq} (370 Bq \cdot kg $^{-1}$) [20]. The calculated external hazard index values were found to vary between 0.55 and 0.86 with average value of 0.728 these values are less than unity, which is 27.2% less than recommended value. The calculated internal hazard index values were found to vary between 0.63 and 1.034 with average value of 0.8584, which is 14.16% less than recommended value [30].

Radioactivity level index: Radioactivity level index ranged from 1.531 to 2.395 Bq \cdot kg $^{-1}$ with average value is 1.9896 Bq \cdot kg $^{-1}$ (Table 2). The average value of Radioactivity level index is higher than the recommended safe limit ≤ 1 [33]. Therefore, the soils have radiation hazard and are harmful to society living there.

Conclusion

The radionuclides in the samples were determined using gamma ray spectrometry. The measured average activity concentrations for ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs in Bq/Kg are 46.93, 110.6, 847.65 and 2.43, respectively. The average activity concentrations of ^{238}U , ^{232}Th and ^{40}K were found to be higher than the world average 35 Bq/Kg, 30 Bq/Kg and 400 Bq/Kg. These results can be considered as base line monitoring for natural background radioactivity levels.

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