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RESEARCH ARTICLE

DETERMINATION OF RADIOACTIVITY LEVELS AND RADIATION HAZARDS IN SOIL SAMPLES COLLECTED FROM HASASA, TOGA AND BISHAN GURACHA, ETHIOPIA

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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 46.708, 97.54, 795.414 and 2.23Bq/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, Dr, Din, Dout, AEDE, Hin, Hex, and I_γ of natural radio-nuclides were determined to be as follow 191.7643, 148.5117, 1.020513, 0.182235, 1.202748, 0.794464, 0.668224 and 1.817106 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.

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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 radio- nuclides 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 knowledge of radionuclides distribution and radiation levels in the environment is important for assessing the effects of radiation exposure due to both terrestrial and extraterrestrial sources. Natural background radiation is of terrestrial and extraterrestrial origin. Terrestrial radiation is due to radioactive nuclides present in varying amounts in soils, building materials, water, rocks and atmosphere. Some of these radionuclides from these sources are transferred to man through food chain or inhalations, while the extraterrestrial radiation originates from outer space as primary cosmic rays (3-6). Studying the levels of radionuclide distribution in the environment provides essential radiological information [7]. 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 [8]. 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 [9]. Many studies world- wide have measured the activity concentration of natural radionuclides in soil to ascertain the levels of contamination [7, 8-14]. The aim of this study is to determine natural (238U, 232Th, 40K) and arti0icial (137Cs) radioactivity levels in soils collected from different points in Hasasa and Toga, Ethiopia. Also, Raeq, D, Hex, and AEDE which will be determined later have been calculated and compared with the results in literature.

MATERIALS AND METHODS

Sample Collection and Sample Processing

A total of 8 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 5cm. 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 [15]. The samples were dried, sieved, packed in 410 gm 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 [16].

Theoretical Calculations

The Activity Concentration

The activity concentrations of the radionuclides in the measured samples were computed using the following relation [16]:

$$C = \frac{C_a}{Ix\varepsilon_{ff}xM_s}$$

Where *Ca* 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 [16].

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, *Dr*. The values of *Dr*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 [17, 18].

$$D_r(nGyh^{-1}) = 0.427xAU + 0.662xATh + 0.043AK$$

Eq. (3) was modified to include the contributions of artificial radionuclides of cesium, (^{137} Cs), as well as cosmic radiation via the following equation [17].

$$D_r(nGyh^{-1}) = 0.427xAu + 0.662xATh + 0.043xAK + 0.03XACs + 34$$

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 [19]. 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 *Dr* 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, *Dout*; the annual effective indoor doses, *Din*; and total annual effective doses, *Dtot*, were calculated according to the following equations [20].

$$D_{out}(mSvy^{-1}) = D_r(mGyh^{-1})x24hx365.25dx0.2x0.7SvGy^{-1}x10^{-6}$$

$$D_{in}(mSvy^{-1}) = D_r(mGyh^{-1})x24hx365.25dx1.4x0.8x0.7SvGy^{-1}x10^{-6}$$

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

The external and internal hazard index: The external (*Hex*) and internal (*Hin*) hazard index due to the emitted γ-rays of the soil samples were calculated and examined according to the following criterion:

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

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 *Hex* 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 [21]. Values of I_γ can be calculated according to the semi empirical formula [22,23].

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

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.

RESULTS AND DISCUSSION

Detector Characterization

Energy Calibration: The detector calibration was performed using a certified standard reference mixed material ^{139}Ce (166 keV), ^{113}Sn (392 keV), ^{85}Sr (514 keV), ^{137}Cs (662 keV), ^{88}Y (898 and 1836 keV) and ^{60}Co (1173 and 1332 keV) in the

energy range (166 - 1850) keV [16]. 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.

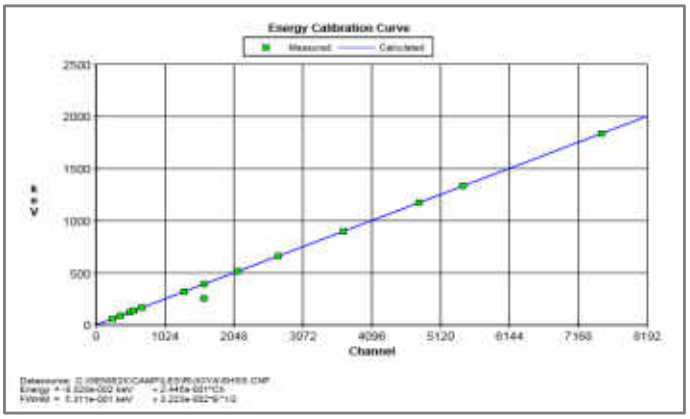

Figure 1. Energy calibration relation between channel numbers and corresponds to energy

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 [24]. 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 ^{226}R in order to cover the range from 60 up to 2000 keV [16]. The graph between efficiency and the energy was plotted.

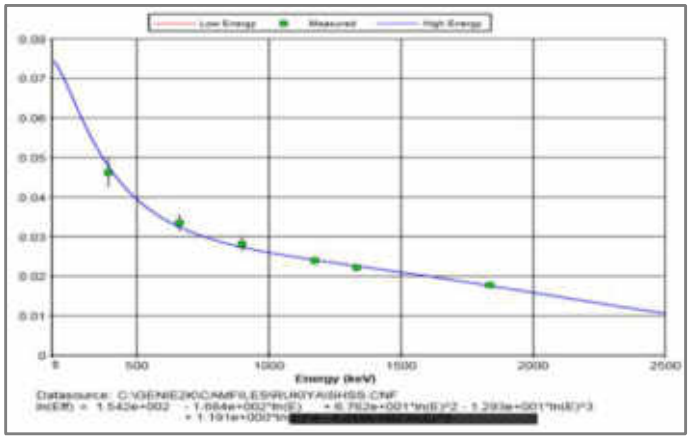

Figure 2. Detector Energy Vs Efficiency Curve

The 295.21 and 351.92 keV of ^{214}Pb and 609.31, 1120.29 and 1764.49 keV of ^{214}Bi gamma ray lines were used to determine the ^{238}U activity concentration. The ^{232}Th activity concentration was determined using 77.11 and 238.63 keV of ^{212}Pb, 911.21 and 968.97 keV of ^{228}Ac gamma lines. The activities of ^{40}K determined directly from the 1460.8 keV gamma lines, respectively. The net count rate under the most prominent photo peaks of all radionuclides 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 [25]. As we can be seen from Table 1, the value of measured activity concentration of ^{238}U, ^{232}Th and ^{40}K were varies from 24.1226 to 56.3, 67.904 to 115.423 and 534.98 to 918.545 Bq/Kg respectively. The average activity concentration of ^{238}U, ^{232}Th and ^{40}K was 46.71, 97.54, 795.464 Bq/Kg, respectively. These values were higher than the recommended world average values for ^{238}U, ^{232}Th, ^{40}K , respectively which are 35, 30 and 400 Bq/Kg for ^{238}U, ^{232}Th, ^{40}K respectively

Table 1. The measured Activity concentration level of natural radionuclides in the soil samples collected from different sites located in Aje, Bishan Guracha, Hasasa, and Toga

	Activity Concentration			
Sample ID	**^{238}U**	**^{232}Th**	**^{40}K**	**^{137}Cs**
ARDVS	30.6 ±2.109688	81.0535 ±6.4536	607.876 ±27.9023	……
DB	56.3 ±3.90955	105.9847 ±8.734	846.001 ±40.033	0.30377 ±0.496669
SA1	48.8 ±3.3	113.93 ±9.57	881.096 ±40.1164	7.40777 ±0.0644194
SBGA-22	52.4 ±3.4934	99.4 ±6.467	908.5 ±41.4039	3.40654 ±0.237108
TSA1	52 ±3.554518	96 ±7.76264	776.3 ±36.2528	2.41325 ±0.181884
TSA2	55.94221 ±3.8107	115.4258 ±9.6338	918.545 ±42.3887	0.586469 ±0.0784295
TSA3	53.51756 ±3.7118	100.6605 ±5.77827	890.412 ±41.028	2.5377 ±0.185931
TSA4	24.1226 ±1.7042	67.90438 ±5.17216	534.982 ±23.6669	1.20966 ±0.096955

The Activity Concentration

The results of analysis of activity concentration of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides in soil samples for different locations of the study area are presented in (Table 1). The activity concentrations of ^{2238}U, ^{232}Th, ^{40}K and ^{137}Cs 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^{60}Co 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.

It is also observed that the measured activity concentration of ^{40}K 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 ^{40}K activity. The activity concentrations of the artificial radionuclide ^{137}Cs 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 ^{137}Cs in all collected soil samples were found to range from 0.30Bq kg^{-1}to 7.4Bq kg^{-1}with an average value of 2.233Bq kg^{-1}. The minimum activity concentration value of ^{137}Cs was obtained

for a soil sample collected from Hasasa, whereas the maximum value was measured in a soil sample collected from Aje 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 ^{137}Cs 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 Raeq, Dr, Dout, Din, Hin, Hex and I_γ of the collected soil samples are shown in table 2.

Radium equivalent activity

It's calculated through the following relation:

Raeq= CRa+1.43CTh + 0.07Ck

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 *Raeq* 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[16].

The values of radium equivalent for different soil samples in area under investigated were calculated by using equation above these values presented in table2 and values ranged from 124.96 to 227.4259 Bq/kg with average value 191.76.27Bq/kg which is lower than the recommended maximum value 370 Bq/kg [16].

Absorbed dose rate in air

The calculated values of *Dr* were found to vary from 96.64 to 173.8nGy h^{-1}, with an average value of 148.5117nGy 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}[17, 18]. Such locations are not safe 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 *Dout*, *Din*, and *Dtot* averages were respectively 0.1822, 1.0205, and 1.202 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 [29].

The external and internal hazard index

The external (*Hex*) and internal (*Hin*) 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·y^{-1}. The maximum values of *Hex* equal to unity correspond to the upper limit of *Raeq* (370 Bq·kg^{-1}) [30]. The calculated external hazard index values were found to vary between 0.43 and 0.78 with average value of 0.668 these values are less than unity, which is 33.2 % less than recommended value. The calculated internal hazard index values were found to vary between 0.5 and 0.93 with average value of 0.79, which is 11.0% less than recommended value [22].

Radioactivity Level Index

Radioactivity level index ranged from 1.19 to 2.13 Bq.kg^{-1}with average value is 1.817 Bq. kg^{-1} (Table 2). The average value of Radioactivity level index is higher than the recommended safe limit ≤ 1 [31]. Therefore, the soils have radiation hazard and are harmful to society living there.

Table 2: The associated radiological hazard parameters indices Raeq, Hex,Hin, Dr and I γ of natural radionuclides

Sample ID	Req(Bq/Kg)	Dr(nGy/h)	Din(mSv/y)	Dout(mSv/y)	AEDE(mSv/y)	Hin	Hex	I γ
ARDVS	150.7566	92.64577	0.636625	0.113683	0.750308	0.604717	0.522015	1.419751
DB	213.7734	164.5859	1.130969	0.201959	1.332928	0.889397	0.737235	1.999134
SA1	217.8876	168.3689	1.156964	0.206601	1.363565	0.886848	0.754956	2.052031
SBGA-2	200.9015	161.3451	1.1087	0.197982	1.306682	0.855904	0.714283	1.949
TSA1	194.7141	153.2092	1.052793	0.187999	1.240792	0.81313	0.67259	1.8242
TSA2	227.4259	173.7961	1.194258	0.21326	1.407518	0.93892	0.787731	2.139288
TSA3	203.6874	161.8499	1.112168	0.198601	1.31077	0.863011	0.718389	1.956946
TSA4	124.9681	112.2925	0.771629	0.137791	0.90942	0.50378	0.438591	1.196498

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 46.708, 97.54, 795.414 and 2.23, 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, 30Bq/Kg and 400Bq/Kg. These results can be considered as base line monitoring for natural background radioactivity levels.

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