Investigations of Natural Radioactivity Levels and Assessment of Radiological Hazard of Tea Samples Collected from Local Market in Ethiopia

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Abstract In this study, the activity concentrations of natural (238U, 232Th, 40K) and artificial radionuclides (137Cs) were measured in six different brands of tea samples. The samples have been collected from local market in Ethiopia (Addis Ababa city) and analysis was done by a detection system consisting of High Purity Germanium (HPGe) detector and multichannel analyser. The average activity concentrations of 238U, 232Th and 40K were found to be 3.66±0.47, 18.28±2.4 and 582.59±29.64 Bq/kg respectively. The activity concentration of 40K is increasing faster than the other NORM for Tea. In order to evaluate the radiological hazard of the natural radioactivity, radium equivalent (RaEq), the external hazard index (H_e), internal hazard index (H_i), the total absorbed dose rate (D), and the annual effective dose equivalent (AEDE) have been calculated and compared with the international acceptable values.

Keywords Natural Radioactivity, Tea sample, Radiological indices, HpGe detector

1. Introduction

Radioactivity is part of the physical environment. The sources of the background radiation in the environment are radionuclides of primordial origin (40K, 238U and 232Th); cosmogenic radionuclides (3H, 7Be, 14C, 22Na) and anthropogenic radionuclides (137Cs, 90Sr, 85Kr). These radionuclides exist in the soil, rocks, water and air.

Studies indicate that about 87% of the radiation dose received by humankind is from the natural radiation of primordial and cosmogenic origin, and the remaining is due to anthropogenic radiation [6]. This dose, to the general population, is received from the natural radiation either by external exposure or internal exposure that may occur via complex environmental path ways [4].

Radionuclides such as 40K, 210Po and 226Ra that occur naturally in soil are incorporated metabolically into plants and ultimately find their way into food and water. Artificial radionuclides behave in a similar manner, and worldwide contamination of the food chains by radionuclides produced during tests of nuclear weapons in the atmosphere has taken place since the 1960s. In addition to root uptake, deposition on foliar surfaces can occur directly or as a result of dry or wet deposition, in which case contaminants can be absorbed metabolically or, more likely, transferred directly to animals that consume the contaminated foliage [Eisenbud].

Tea plants may be subjected to direct and indirect contamination of radionuclides from progenies of 238U and 232Th decay chains or 40K. Radionuclides from fallout can also contaminate tea plants. These radionuclides can be distributed in different parts of the plants according to the chemical characteristics and parameters of the plants and soil [5]. For assessing the effects of radiation exposure due to both natural and artificial radioactivity, part of which dealing with radionuclides of fallout origin. 137Cs and 90Sr is reported by [4].

The aim of this study is to determine natural radioactivity levels (238U, 232Th, 40K) and artificial (137Cs) radioactivity levels in tea samples collected from local market in Ethiopia. The average radium equivalent activity (RaEq), the external hazard index (Hex), the total absorbed dose rate (D), and the annual effective dose equivalent (AEDE) have also been calculated and compared with the results in literature.

2. Materials and Methods

Six samples of different brands of tea have been collected from the local market in Ethiopia (Addis Ababa city) during the year of the experiment. Ethiopian tea brands are originating from two plantations, namely Wush Wush Tea Plantations and Gumaro tea plantation.

The Wush Wush Tea Plantation is located in the Kaffa
zone of Southern Region (460km South West of Addis Ababa) at 1900m above sea level. The soil there is fertile, good drainage, red brown color, reach in organic matter.

THE Gumaro Tea Plantation is located in Illubabor Zone, Oromaya Region, South Western Ethiopia (637 km South West of Addis Ababa) at an altitude of 1718 m. The soil profile is fertile is the same as that of Wush wush plantation, good drainage, red brown color, reach in organic matter.

All brands collected from the Addis Ababa markets were harvested and processed from the tea farms in Gumaro or Wush wush. After collecting, the samples were dried in a drying, In order to remove moisture, tea samples oven at 150°C until constant mass was obtained. Then samples were sealed in standardized Marinelli beaker of volume 0.5 litre. Samples were kept for 30 days for secular equilibrium with 226Ra decay products before the measurements.

In this study, gamma spectroscopy analysis system was used to determine activity concentrations and radiological effects. The system consisted of a coaxial p-type high-purity germanium detector that was linked to a multichannel analyser consisting of analog-to-digital converter and Genie 2000 multichannel analyser software of 8192 channels. The spectra of all samples were perfectly analysed by using Genie-2000 spectra analysis software to calculate the concentrations of 238U, 232Th and 40K [2]. The spectra were analysed for energies of daughter nuclides 214Pb(295.2keV), 214Bi (351.9keV), 214Bi(609.3keV), 214Bi(1120.2keV), 214Bi (1764.4keV) and 214Pb(238.6keV), 208Tl(583.1keV), 228Ac(911.2keV), 228Ac(968.9keV) respectively were recorded. The activity concentrations of 40K were determined directly by measurement of the gamma-ray transitions at 1460.8keV.

2.1. Activity Concentration

The activity concentration of each radionuclide in the sample was determined by using the net count rates (cps) for the same counting time under the selected photo peaks, weight of the sample, the photo peak efficiency, and the gamma intensity at a specific energy as given by the following equation [1].

\[
A = \frac{N \times 100 \times 1000}{P \times E \times W} \tag{1}
\]

where, \(N\) = Net counts per second (c.p.s)
\(P\) = (Sample c.p.s) - (Background c.p.s)
\(P\gamma\) = Transition probability of gamma ray
\(E\) = Efficiency in percent
\(W\) = Weight of the sample in kg.

2.2. Radiological Effects

Since, the distribution of 238U, 232Th and 40K in the environment is not uniform so that with respect to the radiation, the radiological effect of the radioactivity can be measured via absorbed dose(nGy/h). The annual effective dose Equivalent .radium equivalent activity (in Bq/Kg), and external and radiation hazard indices.

2.3. Absorbed Dose (D(nGy/h))

The assessment of radiological hazard due to exposure of external terrestrial gamma-ray radiation from 226Ra(238U), 232Th and 40K in plant, food, rocks, soils and building materials can also be measured using Absorbed Dose Rate in Air (ADRA) at about 1m above the ground. It is computed based on the following equation 3 [7].

\[
D = 0.462U_A + 0.604Th_A + 0.0417K_A \tag{2}
\]

Where the dose rate, D is in nGy/h and \(U_A\) stands for activity of U-238in Bq/Kg, \(Th_A\) stands for activity of 232Th and \(K_A\) stands for activity of 40K. .. This dose rate indicates the received dose from radiation emitted by radionuclides in environmental materials [5].

2.4. The Annual Effective Dose Equivalent (AEDE)

The annual effective dose Equivalent (AEDE) was calculated by using equation 4

\[
AEDE(Sv/y) = D \times DCF \times OF \times T \tag{3}
\]

where D is absorbed dose rate (nGy/ h), DCF is dose conversion factor (0.75Sv/Gy), OF is occupancy factor (0.2), T is the time (8760h/y) [7].

2.5. Radium Equivalent Calculation (R_{eq})

The specific activity of materials containing different amounts of 238U, 232Th and 40K according to [1] was calculated.

\[
R_{eq} = U_A + 1.43Th_A + 0.077K_A \tag{4}
\]

where \(U_A\), \(Th_A\) and \(K_A\) are the activity concentrations of 238U, 232Th and 40K in Bq/kg respectively. The permissible maximum value of the radium equivalent activity is 370Bq/kg.

2.6. External (H_{ex}) and Internal (H_{in}) Hazard Index

The natural radioactivity of tea can be estimated using internal and external radiation indices based on the following expressions [7].
\[ H_{ex} = \frac{U_A}{370} + \frac{Th_A}{279} + \frac{K_A}{4810} \] (5)

\[ H_{in} = \frac{U_A}{185} + \frac{Th_A}{259} + \frac{K_A}{4810} \] (6)

For safe use of material in the tea sample, the external index \((H_{ex})\) and internal index \((H_{in})\) should be each less than unity. Where: \(U_A\), \(U_{Th}\) and \(K_A\) are the activity concentrations in Bq/kg of \(^{238}\)U, \(^{232}\)Th and \(^{40}\)K respectively.

3. Results and Discussion

The results of activity concentrations in the Six samples of different brands of tea have been collected from the local market in Ethiopia (Addis Ababa city) are given in table 1 for the natural radionuclides of \(^{238}\)U, \(^{232}\)Th and \(^{40}\)K. Radium equivalent activity \((Ra_{eq})\), external hazard index \((Hex)\), internal hazards index \((Hin)\), absorbed dose rates \((D)\), annual effective doses equivalent \((AEAD)\) are given in table 2.

| Sample Id | \(^{238}\)U (Bq/kg) | \(^{232}\)Th (Bq/kg) | \(^{40}\)K (Bq/kg) |
|-----------|-----------------|-----------------|-----------------|
| Tea1      | 3.78±0.446      | 19.59±2.5       | 565.77±23.69    |
| Tea2      | 2.88±0.98       | 15.55±2.1       | 634.11±32.21    |
| Tea3      | 3.66±0.48       | 18.53±2.59      | 518.93±28.18    |
| Tea4      | 3.91±0.49       | 18.27±2.6       | 573.00±31.08    |
| Tea5      | 3.67±0.42       | 18.27±2.4       | 624.81±32.77    |
| Tea6      | 4.06±0.46       | 19.48±2.4       | 624.81±32.77    |
| Average   | 3.66±0.47       | 18.28±2.4       | 582.59±29.64    |

Table 2. Radium equivalent (Bq/kg), external hazard index, internal hazard index, absorbed dose rates(nGy/h), annual effective dose equivalent (µSv/y)in tea samples

| Sample Id | Ra_{eq} | Hin | Hex | D   | AEAD |
|-----------|---------|-----|-----|-----|------|
| Tea1      | 75.35   | 0.2 | 0.2 | 38.14 | 46.77 |
| Tea2      | 73.95   | 0.2 | 0.2 | 38.18 | 46.82 |
| Tea3      | 75.38   | 0.18| 0.18| 35.42 | 43.43 |
| Tea4      | 74.16   | 0.2 | 0.19| 37.84 | 46.40 |
| Tea5      | 77.87   | 0.2 | 0.2 | 39.81 | 48.82 |
| Tea6      | 76.49   | 0.2 | 0.2 | 38.76 | 47.53 |
| Average   | 75.53   | 0.19| 0.19| 38.02 | 46.62 |

The activity concentration of the \(^{238}\)U in tea samples ranged from 3.66±0.48 Bq/kg to 4.06±0.46 Bq/kg with an average value of 3.66±0.47 Bq/kg. The estimated average values of \(^{238}\)U in these study are lower than the recommended maximum value of 35 Bq/kg [7].

The activity concentration of the \(^{232}\)Th in tea samples ranged from 15.55±2.1 Bq/kg to 19.59±2.5 Bq/kg with an average value 18.28±6.9 Bq/kg. The obtained value of \(^{232}\)Th is lower than the acceptable value of concentration for \(^{232}\)Th 40Bq/kg [7].

The activity concentration of the \(^{40}\)K in tea samples varied from 518.93±28.18 Bq/kg to 634.11±32.21 Bq/kg with an average value of 582.59±29.62 Bq/kg. The values are greater than the allowable value 400 Bq/kg [7].

\(^{137}\)Cs does not exist in tea sample naturally. The values of radium equivalent activity \((Ra_{eq})\) in tea samples varied from 73.95 Bq/kg to 77.87 Bq/kg with an average of 75.53 Bq/kg. The estimated average values of \(Ra_{eq}\) which is far below the internationally accepted value 370 Bq/kg [7].

The absorbed dose rates due to these radioactive nuclides in tea samples have been found to vary from 35.42 nGy/h to 39.81 nGy/h with an average value of 38.02 nGy/h. These measured values are less than the world average value 60 nGy/h. Annual effective dose equivalent have been calculated from 43.43 µSv/y to 48.82 µSv/y with an average value 46.62 µSv/y respectively which is less than the annual dose limit 1 mSv.

The values of external hazard index, \(Hex\) range from 0.18 to 0.20 and average value was found to be 0.19 for the tea samples. The maximum value of \(H_{ex}\) must be less than unity. All values estimated of \(H_{ex}\) in these study are lower than unity and the values of internal hazard index, \(H_{in}\) range from 0.18 to 0.21 and average value was found to be 0.19 for the tea samples. The maximum value of \(H_{in}\) must be less than unity. All values estimated of \(H_{in}\) in these studies are lower than unity. The values were less than unity in all the samples that indicate the non-hazardous value for human being. Artificial radionuclide like \(^{137}\)Cs was not found in any samples in these studies.

As can be seen from table 3, the range of activity concentration of \(^{238}\)U in Ethiopian tea samples is the same as that measured in Turkish and Indian samples. The samples from Bangladesh show about eight fold concentration of \(^{238}\)U.

\(^{232}\)Th concentration in Ethiopian tea sample is significantly higher than that of Turkish and Indian teas samples. It is nearly half of the concentration observed in Bangladesh tea samples.

\(^{40}\)K in Ethiopian tea samples is comparable with that observed in Turkish tea samples.

4. Conclusions

The activity concentrations and the radiological hazard were calculated from measurements made on the six samples. The results of the study led to the following conclusions.

The mean activity concentration of \(^{238}\)U, \(^{232}\)Th and \(^{40}\)K were indented with average values of 3.66±0.47Bq/kg, 18.28±2.4 Bq/kg and 582.59±29.64 Bq/kg respectively.

Annual effective dose associated with the tea samples have been found to vary from 43.43 µSv/y to 48.82 µSv/y with an average value 46.62 µSv/y respectively. Which is less than the effective dose limit 1 mSv. Values of radium equivalent activity in all samples are less than the permissible maximum value of the radium equivalent activity which is 370Bq/kg according to UNSCEAR 2000 report. The obtained mean values of internal and external hazards index are less than unity.
hazard indices for different tea samples are less than unity. No artificial radionuclide was found in any of the tea samples. Most of the values were less than with other study in the world. So, tea consumption in Ethiopia is non-hazardous for public health.

Table 3. Comparison with Activity Concentration of U-238, TH-232 and K-40 in (Bq/kg)

| Country       | Sample No | 238-U  | TH-232 | K-40  |
|---------------|-----------|--------|--------|-------|
| Turkish tea samples | 1         | 2.8    | 3.8    | 506.60|
| Bangladesh tea   | 2         | 3.1    | 4.2    | 435.20|
| Indian tea sample | 3         | 3.6    | 4.8    | 484.00|
| Ethiopian Tea sample | 4         | 4.1    | 4.3    | 586.40|
|                | 5         | 3.7    | 3.8    | 476.20|
|                | 6         | 3.2    | 2.7    | 432.50|
|                | 7         | 2.7    | 3.2    | 520.60|
|                | 8         | 3.1    | 5.4    | 428.70|
|                | 9         | 3.5    | 4.2    | 452.00|
|                | 10        | 2.8    | 3.2    | 514.00|

**Average**

| Country       | Sample No | 238-U  | TH-232 | K-40  |
|---------------|-----------|--------|--------|-------|
| Bangladesh tea   | 1         | 19.53  | 33.21  | 89.57 |
| Indian tea sample | 2         | 20.78  | 34.93  | 93.28 |
|                | 3         | 23.31  | 37.95  | 102.07|
|                | 4         | 20.97  | 21.24  | 92.62 |
|                | 5         | 22.33  | 34.46  | 92.77 |
|                | 6         | 21.04  | 33.86  | 91.01 |
|                | 7         | 22.18  | 36.92  | 98.13 |
|                | 8         | 22.75  | 37.26  | 100.43|
|                | 9         | 20.65  | 21.24  | 92.71 |
|                | 10        | 20.73  | 33.57  | 89.22 |

**Average**

| Country       | Sample No | 238-U  | TH-232 | K-40  |
|---------------|-----------|--------|--------|-------|
| Indian tea sample | 1         | 2.04   | 1.68   | 847.30|
|                | 2         | 2.53   | 1.29   | 794.76|
|                | 3         | 0.53   | 1.62   | 710.40|
|                | 4         | 2.12   | 1.53   | 759.20|
|                | 5         | 2.68   | 1.15   | 799.94|
|                | 6         | 2.03   | 4.27   | 678.95|
|                | 7         | 2.11   | 1.86   | 676.00|
|                | 8         | 3.33   | 4.80   | 742.59|
|                | 9         | 3.62   | 2.26   | 560.90|
|                | 10        | 2.40   | 1.93   | 929.07|

**Average**

| Country       | Sample No | 238-U  | TH-232 | K-40  |
|---------------|-----------|--------|--------|-------|
| Ethiopian Tea sample | 1         | 3.78   | 19.59  | 565.77|
|                | 2         | 2.88   | 15.55  | 634.11|
|                | 3         | 3.66   | 18.53  | 518.93|
|                | 4         | 3.91   | 18.27  | 573.00|
|                | 5         | 3.67   | 18.27  | 624.81|
|                | 6         | 4.06   | 19.48  | 578.93|

**Average**

| Country       | Sample No | 238-U  | TH-232 | K-40  |
|---------------|-----------|--------|--------|-------|
|                | 1         | 3.66   | 18.28  | 582.59|

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