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Estimation of Activity Concentration of ^{232}Th, ^{137}Cs, and ^{40}K in Soil in Bangladesh

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Abstract This study deals with the assessment of natural radioactivity due to ^{232}Th, ^{137}Cs and ^{40}K in soil samples in Bangladesh. High purity germanium (HPGe) detector based spectrometry system coupled with Multi Channel Analyzer (MCA) was used to obtained gamma-ray spectra. The spectra of all the measurements were saved in the MCA software. MAESTRO for Windows Model A65-B32 containing a software version 5.30 enabled us to obtain the total and net areas under the photo-peaks of the specific gamma-ray energies of interest. The contents of ^{232}Th was determined using 510 keV, 583.14 keV & 860 keV of ^{208}Tl and 911.07 keV & 969.19 keV of ^{228}Ac. The ^{40}K and ^{137}Cs are estimated from 1460 and 662 keV respectively. The mean concentrations of ^{232}Th, ^{40}K and ^{137}Cs were found to be 49.964 ± 5.153, 719.469 ± 47.893 and 7.759 ± 0.717 Bq/kg respectively. The obtained results are compared with the reported and recommended values from other countries and international organizations respectively. The results indicates that there was no fallout in Bangladesh due to nuclear accident like Three Mile Island (1978), Chernobyl (1986) and Fukushima Daiichi (2011) and the low level activity of ^{137}Cs fallout pauses hardly any radiation hazard to the population. The widespread distribution of ^{40}K in soil provides principal contribution to radioactivity in the environment. The study provide a base line data for further study of radiation hazards in Bangladesh in comparison with world average as well as to study the impact of nuclear accident throughout the world.

Keywords MCA, HPGe, Natural radioactivity, Effective dose, Radionuclides etc

1. Introduction

The environmental contamination with radioactivity may cause of human activities like uses of radioisotope for treatment/diagnosis, nuclear weapons and nuclear energy [1,2]. The nuclear reactor accident in Three Mile Island (1978), Chernobyl (1986) and Fukushima Daiichi (2011) also may cause of pervasive of radionuclide into environment and subsequent fallout into soil because of rainfall. In most cases the radioactive wastes are stored with safety criterion but the comprehensive fallout from nuclear weapons/nuclear accident is the potential threat for human being and is the subject of further research. Soil contamination with radionuclide like ^{137}Cs and ^{232}Th may be elevated from fallout of nuclear accident or testing of nuclear weapons. Both of these radionuclide have introduced a substantial amount of radiation into the environment and have caused environmental contamination for long time since their half-lives are long [3]. Such a long lived radionuclides in soil easily uptakes by plants and redistributed in food chains, which would be a potential threat if the radiation exposure exceeds the permissible limit and may be environmental threat to public health [4]. It has been predicted that, 79% of the radiation to which individuals are exposed is from natural sources, and remaining 21% are from man-made activities [5]. According to UNSCEAR (2000), an average radiation dose of 0.29 mSvyr^{-1} is received worldwide via ingestion of natural radionuclides of ^{238}U and ^{232}Th series and ^{40}K during habitual consumption of food and water. This small amounts of ingested or inhaled radioactivity may produce a damaging effect and can become a serious health risk [6]. In addition natural and artificial radioactivity in building materials (bricks and clay) and soils preeminent the potential health hazards for the dwelling population [7-11]. In spite of having difficulties, the nuclear power industry is expanding to contribute power consumption and to reduce the consumption of fossil fuel to restrict the global warming. With ever increasing population, the uses of radioisotopes in medicine also are sky rocketing. Thus the release of radioactive wastes from medicine, industry and research, nuclear weapon testing and radioactive wastes from nuclear reactors into environment are increasing [12]. Since the route of release of radionuclides from nuclear industry is inevitable and cause of environmental contamination, its

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assessment is important for remediation for radiological safety. The natural occurring nuclides of the uranium, thorium and actinium series have to be invaluable indicators of dynamic process in the aquatic environment [13] and a framework against which the introduction of their alpha emitting nuclides can be assessed. For the assessment of effective dose equivalent to the population of Bangladesh, it is necessary to estimate the concentration of various radionuclides in the environment, entering in the various organs of the body as a function of time.

2. Materials and Methods

Sample Collection and Preparation

The soil samples were collected from 8 different locations at AECD Campus at depths of 0-2 inch and 3-5 inch. About 300 gm of each samples from each depth and location was collected in separate plastic container with proper identification code. The depths were so chosen to obtain samples from the surface to plough line for the qualitative and quantitative determination of radioactivity. The samples were dried until they attained constant weight. Each dried samples were grounded to ensure homogeneity and transferred to sealable plastic container, marked individually with identification parameters. Then the prepared samples were ready for counting. Standards (Al_2O_3 based ^{226}Ra and IAEA Soil-6) were also prepared in the same way.

Gamma Spectrometry

The energy calibration of the HPGe detector [14] was performed by using known gamma point sources like ^{137}Cs and ^{60}Co with energies 662 keV, 1172 keV and 1332 keV. The energies response linearly with channel numbers and the MCA was adjusted to suitable channel numbers by entering the energies of the calibration source in keV into the MCA to convert all 4096 channels to respective energies. For quantitative determination of the radionuclides in unknown sample, the detector efficiency which relates the number of pulses counted to the number of gamma photons incident on the face of the detector was adjusted [15]. The energies spread in multi-channel analyzer were adjusted to 0-3366 keV energy over 16384 channels. The energies response linearly with channel numbers and the MCA was adjusted to suitable channel numbers by entering the energies of the calibration source in keV into the MCA to convert all channels to respective energies. The detection efficiencies were calculated using Al_2O_3 based ^{226}Ra standard. The content of ^{226}Ra was measured using γ-energy of 295.21 keV & 351.92 keV for ^{214}Pb and 609.31 keV, 1120.3 keV & 1764 keV for ^{214}Bi. The contents of ^{232}Th was determined using 510 keV, 583.14 keV & 860 keV of ^{208}Tl and 911.07 keV & 969.19 keV of ^{228}Ac. The ^{40}K and ^{137}Cs are estimated from 1460 and 662 keV respectively. The daughter products of ^{226}Ra and ^{232}Th in all samples were omitted from our consideration because either their activities were less than the minimum detectable limit or their peaks were of bad shapes.

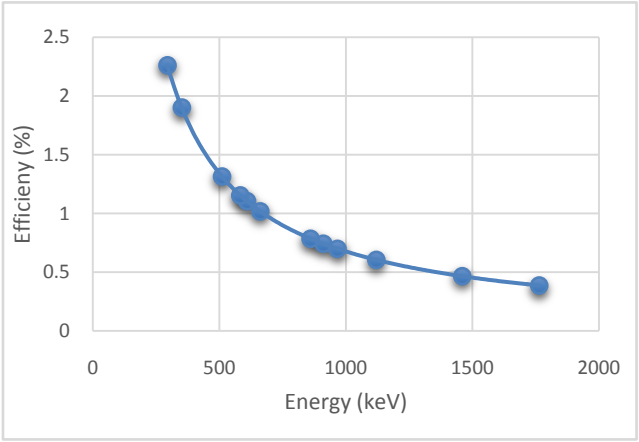

Figure 1. Efficiency Calibration curve

Maintaining the geometrical similarity and constant counting time (e.g. 5000 seconds) the gamma spectrometry was performed and activity was calculated for each samples by using the following formula [14].

$$A_s = \frac{C \times 100}{\varepsilon(E) \times f \times W}[Bq/Kg] \quad (1)$$

Where

C is the count per second

W is the weight of the sample (kg)

ε (E) is counting efficiency of the specific nuclide's energy E (%)

f is the absolute transition probability of gamma decay through the selected energy as for E.

With the knowledge of the statistical behavior of error in observation i.e. deviation from normal value, the error calculation was done by the following formula [16]

$$\sigma = \sqrt{\frac{S+B}{T^2} + \frac{B}{T^2}} \quad (2)$$

Where (S+B) is the total sample and background counts

B is the total background counts

T is the counting time in seconds.

3. Results and Discussion

The natural radioactivity concentrations (average) of ^{137}Cs, ^{232}Th and ^{40}K in different soil samples of Bangladesh are presented in figure-2. For the nuclides of ^{232}Th, measurement were taken on the 911 keV & 967 keV peaks of ^{228}Ac and 583 keV, 511 keV and 860 keV peaks of ^{208}Tl. It is observed that the activity of ^{228}Ac varies from 64.452 ± 6.448 to 83.645 ± 7.079 Bq kg^{-1} with an average of 71.434 ± 6.655 Bq kg^{-1}. The concentration of ^{208}Tl (583, 510 and 860 keV) were found to be in the range from 25.858 ± 3.674 to 31.805 ± 3.684 Bqkg^{-1} with an average of 28.494 ± 3.651 Bq kg^{-1}.

The average ^{232}Th concentration was found to be 49.964 ± 5.153 BqKg^{-1}. The activity of ^{40}K was estimated from 1460 keV peak and found to vary from 683.752 ± 45.943 to 793.123±47.925 Bq kg^{-1} with an average of 719.469 ± 47.893. The maximum and the lowest activity have been found 793.123±47.925 Bq kg^{-1} and 683.752 ± 45.943 Bq kg^{-1} respectively. The average concentration of ^{137}Cs is 7.759 ± 0.717 Bq kg^{-1}. ^{137}Cs is a fission product and could be originated from Nuclear Weapon tests, or Reactor accident. Because of its long half-life (30.17 yrs), ^{137}Cs may accumulate in soil by rain from fallout or from imported foodstuffs such as meat, milk etc.

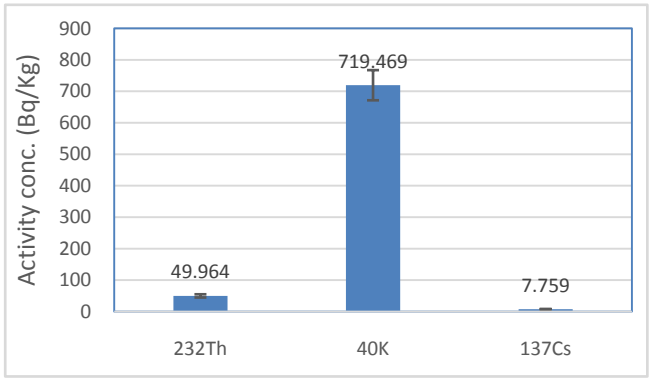

Figure 2. Average Activity conc. of Th, K and Cs

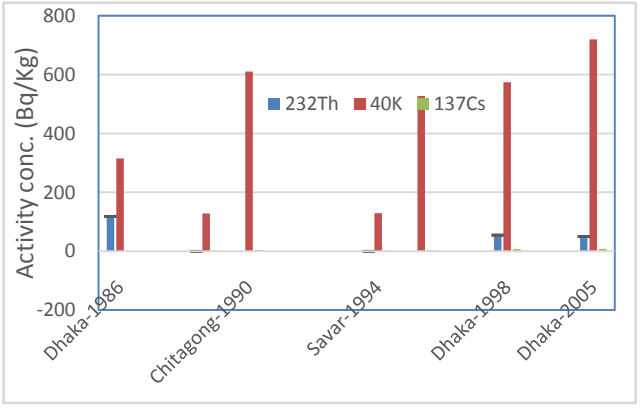

Figure 3. Variation of conc. in distinct years

Figure-3 shows that that the activity concentration of ^{232}Th and ^{40}K and ^{137}Cs are varied randomly in the distinct years. In 1986 the ^{232}Th concentration in soil samples of Dhaka region was 118.8 Bq/Kg which decreased significantly in 1998, reached at 55±14 Bq/Kg and in the 2005 it reduced to 49.964 ±5.153 Bq/Kg. In 1986 the ^{40}K concentration in soil samples of Dhaka region was 315.2 Bq/Kg which increased significantly in 1998, reached at 574±111 Bq/Kg and in the 2005 it reached to 719.749± 47.893 Bq/Kg. The reason of variation is unknown and further study is required. In 1986 the concentration of ^{137}Cs was below detection limit (BDL) which was found 7± 2 Bq/Kg in 1998 and 7.759 ± 0.717 Bq/Kg in 2005 respectively. Further study about this fission product is recommended.

The experimental activity concentration of ^{232}Th and ^{40}K and ^{137}Cs are compared with the world average. In Figure-4 [17-22], it is observed that the ^{40}K activity concentration in Pakistan (646 Bq/Kg), India (716.6 ±7.78 Bq/Kg) and Bangladesh (719.469±47.893 Bq/Kg) are almost similar. Bangladesh has the highest concentration and more than two times conc. that exhibit in the case of Turkey, Greece, Egypt, Iraq and Malaysia. Figure-5 [19-22] shows that ^{232}Th activity concentration in Pakistan (84Bq/Kg) and India (86.6 ±2.62 Bq/Kg) are almost similar but Malaysia has the highest concentration of 108.675±2.6 Bq/Kg. Bangladesh seems to have the lower concentration of 49.964 ±5.153 Bq/Kg but it should not be considered as environmental safe zone near future since it would be the cause of fallout of fission product due to Nuclear accident in Fukushima. The obtained average ^{137}Cs concentration is higher than those observed in Pakistan & Brazil but lower than Poland, Turkey and Hungary as shown in Figure-6(a) [23-26]. In Figure-6(b) [23-27] the ^{137}Cs concentration of Ukraine is found to be nearly 150 times higher than in Bangladesh which would be the cause of Chernobyl nuclear accident.

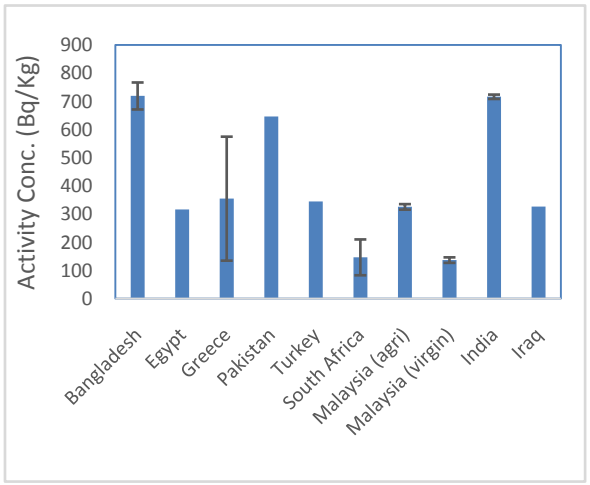

Figure 4. Comparison of K conc. with world average

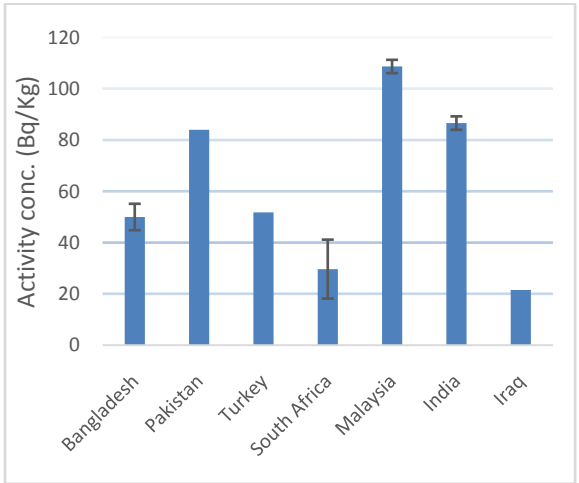

Figure 5. Comparison of Th conc. with world average

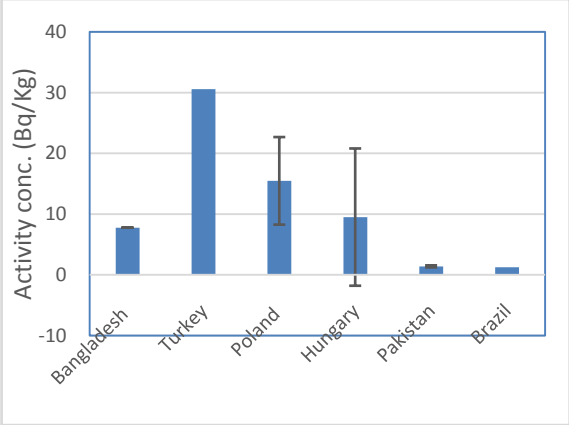

Figure 6(a). Comparison of Cs conc. with world average

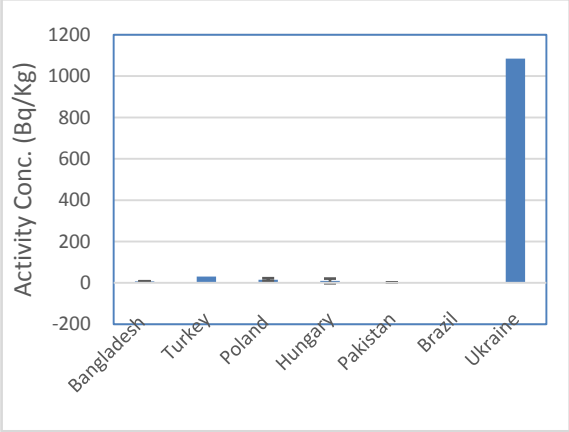

Figure 6(b). Comparison of Cs conc. with world average

4. Conclusions

The natural radioactivity in soil samples were estimated by the gamma spectroscopy and the average radioactivity concentrations of ^{232}Th, ^{40}K and ^{137}Cs in soils are found to be 49.964 ± 5.153, 719.469 ± 47.893 and 7.759 ± 0.717 Bq/kg respectively. The ^{137}Cs activity in soil samples was found to be constant over twenty years and is almost negligible. Therefore, it may be concluded that there was no fallout in Bangladesh due to nuclear accident like Three Mile Island (1978), Chernobyl (1986) and Fukushima Daiichi (2011). Till now low level activity of ^{137}Cs fallout pauses hardly any radiation hazard to the population. The widespread distribution of ^{40}K in soil provides principal contribution to radioactivity in the environment. The results provide a base line data for further study of radiation hazards in Bangladesh in comparison with world average as well as to study the impact of nuclear accident throughout the world.

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