J. Bangladesh Acad. Sci., Vol. 43, No. 2, 141-148, 2019
DOI: https://doi.org/10.3329/jbas.v43i2.45735

ASSESSMENT OF RADIOACTIVITY AND RADIOLOGICAL HAZARD OF DIFFERENT FOOD ITEMS COLLECTED FROM LOCAL MARKET IN BANGLADESH

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ABSTRACT

Sixteen samples in three categories vegetables, cereals (rice, wheat, maize, pulse) and powdered milk were collected from local markets (Dhaka city) in Bangladesh and analyzed by using High Purity Germanium (HPGe) Detector for the assessment of natural and artificial radioactivity. In vegetables (potato, raw banana, giant taro, red amaranth), the average activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K were found to be 37.82±11.57, 54.93±9.98 and 617.43±65.69 Bqkg^{-1} respectively, for cereals (rice, wheat, maize, pulse) 24.01±3.67, 31.46±4.00 and 474.83±27.68 Bqkg^{-1} respectively and for milk samples 15.01±3.65, 26.73±6.77 and 494.21±38.71 Bqkg^{-1} respectively. The average values of outdoor annual effective dose were found to be 92.18, 61.19 and 52.37μSvy^{-1} in vegetables, cereals and milk samples respectively. No artificial radionuclide was found in any of these samples. The average value of radium equivalent activity in all samples was 113.89 Bqkg^{-1} which was less than maximum permissible value 370 Bqkg^{-1}. The values of external hazard indices for vegetables, cereals and milk samples varied from 0.31 to 0.66, 0.17 to 0.40 and 0.11 to 0.36 respectively which is less than unity in all samples that indicate the non-hazardous nature of the samples. The average values of annual effective ingestion dose rate from foods (for adult) were 274.33 μSvy^{-1}, 533.60μSvy^{-1}, and 132.73μSvy^{-1}for ^{232}Th, ^{226}Ra and ^{40}K respectively. These data would be useful to establish a baseline for natural radioactivity concentrations in food items consumed in Bangladesh.

Keywords: Radioactivity, HPGe, Effective dose, External hazard index, Radium equivalent activity, Annual effective ingestion dose rate

INTRODUCTION

Radiation can be described as the energy of the electromagnetic waves which emit or transmit energy in the form of waves or particles through space or through a material medium. Radiations are present in the environment since the big bang occurred. During the last few decades, radioisotopes and nuclear explosion have been contaminated our environment. Humans and their foodstuffs are exposed to various types of natural radiation that are originated from primordial, cosmic, terrestrial, natural decay series radioactive nuclides (Dinh *et al*. 2011). Natural radiation is universally presented and generally accepted source of risk and thus serves as one reference against which to compare the risks of other radiation exposures. Natural background radiation averages about 100 mremyr^{-1}, but much higher levels are encountered in some parts of the world about 2000 mremyr^{-1} (ADA 1996). Study says, approximately 25% (Semin. Nucl. Med.1986) of all food products are lost after harvesting because of insects, vermin, and spoilage. Now-a-days, a significant number of chemicals are used

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on food products for preventing losses and all of these used chemicals contained with great numbers of radio nuclides like as ^{226}Ra, ^{232}Th, ^{40}K etc ionize the foodstuffs and foodstuffs become radioactive. Consuming food containing radionuclides is particularly dangerous. If anyone ingests or inhales a radioactive particle, it continues to irradiate the body as long as it remains radioactive and stays in the body (Doc Alan *et al.* 2011). Children are much more susceptible to the effects of radiation and stand a much greater chance of developing cancer than adults. However, studies on the radioactivity of the consumable foods assume importance as it is necessary to estimate the ingestion dose to the public.

MATERIALS AND METHODS

Sixteen samples of different vegetables, grains, powder milks have been collected during the year of the experiment (2017) from the local market in Bangladesh (Dhaka city). The collected samples were dried properly so that it can be grinded into powder. The powdered samples were then sieved using a fine aperture mesh screen (mesh size 2µm) in order to remove extraneous items like plant material, roots, pebbles etc. and to obtain a fine grained sample that would present a uniform matrix to the detector. Then the powder of the samples was dried at 105°C-110°C until a constant weight was achieved and it was ensured that any significant moisture was removed from the samples. Upon collection, all the samples were properly packed and marked for their identification code. The samples were then transported, stored and processed at the sample preparation laboratory of Atomic Energy Centre, Dhaka. Then the samples were transferred to cylindrical plastic containers of 7 cm height and 5.5 cm in diameter and the weights of the samples were recorded using an electrical balance. The sample-filled plastic containers were sealed tightly with cap and wrapped with thick vinyl tape around their necks; marked individually with identification number and date of preparation and net weight and then stored for about 30 days to assume secular equilibrium between ^{238}U and ^{232}Th series and their daughter progenies. HPGe (High purity germanium) detector was used to measure the radioactivity of the samples. Firstly the background level of germanium detector is determined. The background-level measurement is significant and used to determine the minimum detectable activity. This is particularly important in the case of low-level activity sources (Strom 1958).

Background measurement was counted for the same time duration as for the sample which was 5000 sec. Prior to the measurement of the samples, the environmental gamma background at the laboratory site was determined with an identical empty plastic container using in the sample measurement.

To qualitatively identify the contents of radioactive nuclides in the samples and to quantitatively determine their activities, the gamma spectrometric measurements on samples were carried out using a high-purity germanium (HPGe) detector coupled with DSPEC jr 2.0 Digital Signal Processing Gamma Ray Spectrometer. The effective volume of the detector was 83.469 cm^3 and energy resolution of the 1.33 MeV energy peak for ^{60}Co was found as 1.69 keV at full width half maximum (FWHM) with a relative efficiency of 19.6%. The equal counting time 5000s for background and sample measurement was chosen to minimize the uncertainty in the net counts. The spectrum of each sample was analyzed and the identification of unknown radioactive nuclides was carried out by considering their peak centroid energies. The energy regions selected for the corresponding radionuclides were 295 and 352 keV of ^{214}Pb and 609, 1120 and 1764 keV of ^{214}Bi for ^{226}Ra, 583 and 2614 keV of ^{208}Tl, 911 and 969 keV of ^{228}Ac for ^{232}Th and 1460 keV for ^{40}K.

The centroid energies of the peaks from the spectrum were compared with the reference gamma-ray energies. In this study, the gamma reference sources ^{137}Cs (monoenergetic gamma source), ^{60}Co and ^{40}K were chosen for energy calibration due to a wide range of gamma-ray energies emitted over the entire energy range of interest. These nuclides are contained in the same geometry and placed at the same position in the detector system as that of sample measurement in order to reduce the error in the determination of the peak energy. In order to determine the detector efficiency, a standard source was made by mixing ^{152}Eu of known activity with Al_2O_3 matrix. Using this standard source, the counting efficiency curve of the HPGe detector was drawn and is calculated using the following equation:

$$\text{Efficiency} = \frac{\text{CPS}}{\text{DPS} \times I\gamma} \qquad (1)$$

where CPS = counts per second for the radionuclide present in the standard sample,
DPS = disintegration per second, and
I_γ= γ -ray intensity of the source.

For the calculation of the activity concentration of a particular radionuclide in the measured samples, the number of counts under the full-energy peak areas (corrected for background peak areas), the counting time, the absolute full-energy peak efficiency for the energy of interest and the gamma-ray emission probability corresponding to the peak energy are used. The specific activity of individual radionuclides in samples is given by the following equation:

$$A = \frac{N \times 100 \times 1000}{P_\gamma \times \varepsilon \times W} \qquad (2)$$

Where

A= activity of the sample in BqKg^{-1}

CPS= the net counts per second

background value

= CPS for the sample – CPS for the

E=the counting efficiency of the gamma energy

Pγ = absolute intensity of the gamma ray and W= net weight of the sample (in gm.).

The errors in the measurement have been expressed in terms of standard deviation ($\pm\sigma$), where σ is expressed as,

$$\sigma = \left[{N_s}/{T_s^2} + {N_b}/{T_b^2} \right]^{1/2} \qquad (3)$$

Where, N_s is the net counts measured in time T_s and N_b is the background counts measured in the T_b. The standard deviation ($\pm\sigma$), in cps was converted into activity in BqKg^{-1}. Due to a non-uniform distribution of natural radionuclides in the samples, the actual activity level of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K in the samples evaluated by means of a common radiological index named the radium equivalent activity (Ra_{eq}) (Beretka *et al.* 1985) by the expression:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \qquad (4)$$

where, C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively in Bqkg^{-1}.

The permissible maximum value of the radium equivalent activity is 370 Bqkg^{-1}.

This corresponds to an effective dose of 1mSv for the general public (Ajayi 2009). A direct connection between radioactivity concentrations of natural radionuclides and their exposure is known as the absorbed dose rate in the air at 1 meter above the ground surface. The mean activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K (Bqkg^{-1}) in the samples are used to calculate the absorbed dose rate given by the following formula (Belivermis *et al.* 2010).

$$D = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \qquad (5)$$

where, D is absorbed dose rate in $nGy.h^{-1}$. Then, the external hazard was calculated by the formula:

$$H_{ex} = (C_{Ra}/370) + (C_{Th}/259) + (C_K/4810) \quad (6)$$

The annual effective dose equivalent can be estimated using the following formula (Belivermis *et al.* 2010)

$$AEDE(\mu Svy^{-1}) = D(nGyh^{-1}) \times 8760\ h \times 0.2 \times 0.7 Sv.Gy^{-1} \times 10^{-3} \quad (7)$$

By using a conversion factor of 0.7 $SvGy^{-1}$, which converts the absorbed dose rate in air to human effective dose and 0.2 for the outdoor occupancy factor proposed by the UNSCEAR 2000 report were used.

The annual effective dose D to individuals due to the intake of the radio-nuclides can be estimated using the following equation,

$$D_{eff} = AIE \quad (8)$$

where, D_{eff} = Annual effective dose due to ingestion of food is in Svy^{-1}

A= Activit of radionuclides in food samples is in $BqKg^{-1}$

I= Annual intake of food is in Kgy^{-1}

E= Conversion factor is in $SvBq^{-1}$

The conversion factor E varies with both radioisotope and the age of the individual (ICRP 1990). I depends strongly on a given age group (ICRP 1990). The calculation of the annual ingestion dose has been done for different age groups: such as children (2-7 years, 7-12 years, and 12-17 years) and adult (above 17 years). The dose due to food depends on food consumption. In this study all six category samples (vegetables, rice, wheat, maize, pulses, and powdered milk) are usually consumed regularly in Bangladesh. So, overall by this study it can be measured the total annual effective dose for people residing in Dhaka city in Bangladesh. The value of the annual average consumption of food was taken from the report of Bangladesh Bureau of Statistics 2010 (BBS 2011).

RESULTS AND DISCUSSION

The measured activity of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in all samples collected from local market in Bangladesh are shown in Table1.

The activity concentration of the ^{226}Ra in all samples ranged from 6.38±3.71 to 79.56±26.42 $Bqkg^{-1}$. The highest activity concentration of the ^{226}Ra was found in red Amaranth and lowest activity concentration of the ^{226}Ra was found in powdered milk (Dano). The activity concentration of the ^{232}Th in all samples ranged from 6.88±3.00 $Bqkg^{-1}$ to 75.34±19.42 $Bqkg^{-1}$. The highest activity concentration of the ^{232}Th was found in Red Amaranth and lowest activity

Radiological indices

The radiological parameters such as indices of radium equivalent activity (Ra_{eq}), absorbed dose rate (D), internal hazard index (H_{in}), external concentration of the ^{232}Th was found in powdered milk (Dano). The activity concentration of the ^{40}K in all samples varied from 296.97±23.24 $Bqkg^{-1}$ to 834.30±57.57 $Bqkg^{-1}$. The highest activity concentration of the ^{40}K was found in raw banana and lowest activity concentration of the ^{40}K was found in rice (Minikate).

hazard index (H_{ex}) and annual effective dose equivalent (AEDE) have been measured to estimate the radiological risk due to the presence of ^{226}Ra, ^{232}Th and ^{40}K in the samples. Table 2 depicts the values of Ra_{eq}, D, H_{in} , H_{ex} and AEDE.

The values of radium equivalent activity in all samples have been found to be varied from 40.71 $Bqkg^{-1}$ to 243.29 $Bqkg^{-1}$ with an average of 113.89 $Bqkg^{-1}$ which is far below the internationally accepted value of 370 $Bqkg^{-1}$. Annual effective dose equivalent has been calculated from 24.98μSvy^{-1} to 138.08 μSvy^{-1}

Table 1. The activity concentrations (Bqkg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K in samples.

Sample ID	^{226}Ra (Bqkg^{-1})	^{232}Th (Bqkg^{-1})	^{40}K (Bqkg^{-1})
1. Potato	39.67±3.27	33.26±13.75	363.04±31.28
2. Raw Banana	48.90±6.50	21.35±6.93	834.30±57.57
3. Giant Taro	51.60±3.72	21.33±6.16	545.13±46.03
4. Red Amaranth	79.56±26.42	75.34±19.42	727.24±127.87
5. Rice (Minikate)	20.83±2.10	13.21±2.5	296.97±23.24
6. Rice (Chinigura)	35.28±2.83	20.75±3.86	416.64±28.78
7. Wheat (Red)	15.19±4.00	19.46±3.15	319.32±22.64
8. Wheat (White)	17.38±4.22	30.77±3.74	390.93±24.62
9. Maize	48.10±2.92	25.95±3.68	371.02±25.06
10. Grass Pea	47.56±6.09	32.47±4.2	705.85±32.59
11. Green Gram	23.06±4.90	21.01±3.97	647.91±31.54
12. Red Lentil	29.84±4.52	38.38±4.01	488.87±26.25
13. Bengal Gram	28.25±5.27	23.31±4.05	526.82±30.68
14. Matar Dal	49.07±3.19	14.75±3.53	583.98±31.36
15. Powder Milk (Dano)	6.38±3.71	6.88±3.00	318.05±32.19
16. Powder Milk (Diploma)	47.08±9.83	23.14±4.29	670.38±45.22

Table 2. The Radium equivalent activity, absorbed dose rate, Annual effective dose equivalent, external and internal hazard indices associated with different collected samples.

Sample ID	Radium Equivalent Activity (Ra_{eq}) (Bqkg^{-1})	Absorbed Dose rate (D)	External Hazard Index (H_{ex})	Internal Hazard Index (H_{in})	AEDE (μSvy^{-1})
1. Potato	115.19	53.56	0.31	0.42	65.68
2. Raw Banana	143.67	70.28	0.39	0.52	86.19
3. Giant Taro	124.08	59.46	0.34	0.47	72.92
4. Red Amaranth	243.29	112.59	0.66	0.87	138.08
5. Rice (Minikate)	62.58	29.98	0.17	0.23	36.77
6. Rice (Chinigura)	97.03	46.21	0.26	0.36	56.67
7. Wheat (Red)	67.61	32.09	0.18	0.22	39.35
8. Wheat (White)	91.48	42.92	0.25	0.29	52.63
9. Maize	113.78	53.37	0.31	0.44	65.45
10. Grass Pea	148.34	71.02	0.40	0.53	87.10
11. Green Gram	102.99	50.36	0.28	0.34	61.76
12. Red Lentil	122.36	57.35	0.33	0.41	70.33
13. Bengal Gram	102.15	49.10	0.28	0.35	60.22
14. Matar Dal	115.13	55.93	0.31	0.44	68.59
15. Powder Milk (Dano)	40.71	20.37	0.11	0.13	24.98
16. Powder Milk (Diploma)	131.80	63.68	0.36	0.48	78.10
Average	**113.89**	**54.27**	**0.31**	**0.41**	**66.55**

Table 3. The average annual effective dose rate (mSvy^{-1}) due to ingestion

Age group	**^{232}Th (mSvy^{-1})**	**^{40}K (mSvy^{-1})**	**^{226}Ra (mSvy^{-1})**
2-7 years	0.417	0.449	1.181
7-12 years	0.366	0.278	1.524
12-17 years	0.298	0.162	2.858
17-Adult	0.274	0.132	0.533

Table 4. Comparison of the calculated data of different food items with those of the similar items from other countries.

Country/ Region	Radioactivity Concentration (Bqkg^{-1})			References
Radionuclides	^{226}Ra	^{232}Th	^{40}K	
Bangladesh (Local market)	36.7	26.3	513	Present Work
Saudi Arabia	9.6	6.8	74.5	(Zain 2013)
Iran/France	0.05	142	434	(Barati. H *et al.* 2006)
Brazil	-----	1.7 – 3.7	489	(Appoloni CR *et al.* 2002)
Indian	2.5	1.02	34.4	(Allan G. Gnana *et al.* 2010)
Egypt Elexandria	0.44	-----	47. 3	(Abdelfatah F. *et al.* 2007)
Syria	----	----	129-435	(Almasri MS et al.2004)
Jordan	0.5-2.14	0.78-1.28	349-392	(Anas. M. *et al.* 2010)
Venezuela	---	---	402	(UNSCEAR 2000)

with an average value 66.55 µSvy^{-1} respectively which is less than the recommended value of International Atomic Energy Agency (IAEA) which is 1000 µSvy^{-1}. On the other hand, the values of external and internal hazard indices for different samples varied from 0.11 to 0.66 with the average value of 0.31 and from 0.13 to 0.87 with an average of 0.41. The values were less than unity in all the samples that indicate the non-hazardous for human being.

The annual effective dose rate due to ingestion

The annual effective dose rate due to ingestion of these selected foodstuffs by different age groups of people are calculated and it shows that the obtain values are less than the worldwide excepted safe range of the effective dose rate (Table 3).

The studied data is compared with the data of other countries and is given in the following Table 4.

CONCLUSION

The activity of ^{232}Th, ^{226}Ra and ^{40}K radionuclides were determined for the most available food items consumed in Bangladesh. The measured values were found to be within the worldwide ranges as reported in this literature. The mean values of activity of radionuclides ^{232}Th, ^{226}Ra and ^{40}K of sixteen samples of food items were identified as 26.34 BqKg^{-1}, 36.73 BqKg^{-1} & 512.90 BqKg^{-1} respectively. The concentration values are found to be lower than those of reported in other countries. The present result concludes that the radionuclides level in studied samples for children and adult is below the maximum permitted by IAEA. So, these kinds of food can normally be consumed. The average annual effective doses due to intake were calculated for different age groups, children (2-7years, 7-12years, and 12-17years) and adults (above 17years). However, these values are far below for all age groups within the typical worldwide range of annual dose (200– 800 mSv) due to the ingestion of all natural radiation sources (UNSCEAR 2000). The data of this study reported here will help to establish a baseline for natural radioactivity concentrations in food samples consumed in Bangladesh and help to develop future guidelines in the country for radiological protection for the people.

ACKNOWLEDGEMENT

The authors are very grateful to the office and laboratory staff of Health Physics Division, Atomic Energy Centre (AEC), Dhaka, for their cooperation and providing with laboratory facilities. This research work has been supported by the Jagannath University research fund.

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(Received revised manuscript on 10 November 2019)