Natural Radioactivity Level of $^{238}$U, $^{232}$Th, and $^{40}$K in Baby Food and Committed Annual Effective Dose Assessment in Bangladesh

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Abstract: Worldwide, the contagion of chronic diseases like diabetes and cancer among children has made the nutritionists thoughtful. Food contamination with radioactivity, became a severe health threat for children below five since they are under developing immune systems and cannot fight off infections like adults. Radioactivity concentrated in food chain may be transferred to human body and increase the cumulative risk of developing cancer and some other diseases. Therefore the assessment of radioactivity levels in baby food and their associated doses are of crucial importance for health safety. The study is focused on the radiation contamination of baby food due to the nuclear disasters and make public awareness about infant’s nutrition followed by the health safety. The natural radioactivity level due to $^{238}$U, $^{232}$Th and $^{40}$K in baby food (cereals and powder milk) samples, marketed in Bangladesh were estimated and annual effective dose was assessed. Gamma spectrometry was performed by HPGe detector coupled with MCA, and certified reference materials were used for quantification and quality control. The average concentrations of $^{238}$U, $^{232}$Th, and $^{40}$K were found to be 5.42±0.42, 5.71±0.50 and 334.4±12 Bq.Kg$^{-1}$ in milk sample and 2.98±0.38, 3.94±0.29 and 155.7±7.5 Bq.Kg$^{-1}$ in cereal sample respectively. The committed effective doses due to ingestion of natural radionuclides ($^{238}$U, $^{232}$Th and $^{40}$K) from the consumption of milk and cereals for age group <1 year were 1.30 and 0.51 mSv/yr and for age group 1-2 year were 0.31 and 0.15 mSv/yr respectively. The age group < 1yr, who are completely depend on formula milk are under highly threat since their annual effective dose 1.30 mSv/yr exceeds the recommended permissible limit of 1 mSv/yr. The data generated in this study will provide base-line radiometric values of activity concentration and annual effective dose attributed from baby foods in Bangladesh.

Keywords: MCA, HPGe Detector, NaI (TI) Detector, Natural Radioactivity, Effective Dose, Radio-nuclides

1. Introduction

Food contamination with natural radioactivity, now became a serious health threat for human being. According to UNSCEAR, an average radiation dose of 0.29 mSv yr$^{-1}$ 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 [1]. Even small amounts of ingested or inhaled radioactivity may produce a damaging effect and can become a serious health risk [2]. Children below five are at high risk for foodborne illness and related health complications because they are under developing immune
systems and cannot fight off infections like adults [3]. Thus, food poisoning for this age group can be dangerous and even a small amounts of toxins in baby food can build up chemical contamination over time [4]. Research shows that 95% commercial baby foods are contaminated with toxic heavy elements like lead, arsenic, mercury & cadmium and these trace amounts in food can alter the evolving brain and erode a child’s IQ [3]. Consumption of food which contains radionuclides is particularly dangerous. If an individual ingests or inhales a radioactive particle, it continues to irradiate the body as long as it remains radioactive and stays in the body [5]. Decades of research show clearly that any dose of radiation increases an individual's risk of developing cancer. Radiation levels can be concentrated in the food chain and further consumption adds to the cumulative risk of developing cancer and some other diseases [6]. In Bangladesh the demand for milk and milk products is increasing because of the rapid increase in population and growing nutrition awareness. National milk production can only meet 13% of the current milk consumption [7]. The increasing demand of milk production is met by importing milk powder mainly from abroad and assessment of radioactivity become significant since the Chernobyl disaster exposed to high radiation doses even up to 50 Gy to children in the contaminated areas due to intake of radioactivity contaminated milk produced locally. Several studies have found that the incidence of thyroid cancer among children at Belarus in Ukraine and Russia has risen sharply since the Chernobyl disaster. The International Atomic Energy Agency (IAEA) noted around two thousand documented cases of thyroid cancer in children who were between 0 and 14 years of age when the disaster occurred which is far higher than normal [8]. Assessment of radioactivity levels in powdered milk & cereals and their associated doses are of crucial importance for health safety. In Bangladesh, people are mostly dependent on dry baby food such as cereals and powder milk available in the local market. That is why; locally available ready-made and most commonly used cereals and powder milk are the area of the present study. The aim of the study is mainly focused on the determination about radiation contamination due to the nuclear disasters and make public awareness about infant’s nutrition followed by the health safety.

2. Materials and Methods

2.1. Sample Collection and Preparation

Baby food samples such as cereals, junior’s Horlicks and powdered milks including lactogens and adult milk were collected from local markets in and around Savar urban area near Dhaka city. Although, all samples were collected from Savar areas, they are available all over the country. The samples were dried, approximately 75 gm of each samples were taken into air tight plastic pots having the same geometry and encoded with proper sample identification (ID) numbers accordingly. Then the samples allowed to decay naturally for four weeks before counting to assure the parent-daughter equilibrium in the natural decay series. Several Certified Reference Materials (CRMs) namely NIST-SRMs Coal Fly, CFA 1633b, and IAEA CRMs Soil-7 and Lake Sediment SL-1 were prepared same way like other samples. Comparative standardization approach was performed for quantification of elements in all the prepared samples and standards. The reason of using these CRMs was to determine wide number of elements as well as to control the quality of analysis.

2.2. Gamma Spectrometry

Gamma spectrometry was performed by HPGe detector with relative efficiency of 40% and resolution (FWHM) 1.8 keV at 1332 keV γ-energy of 56Co, coupled with multi-channel analyzer (MCA) and PC based Genei-2000 acquisition software, to measure the natural radioactivity in the samples. HPGe detector is widely used for gamma spectroscopy because of its superior resolution over NaI (TI) detector [9]. The energy calibration of the HPGe detector [10] was performed by using Al2O3 based 226Ra sources and their daughter nuclides 214Pb and 214Bi as known energy sources.

The efficiency of the HPGe detector is determined from the following relation,

\[ \varepsilon(\%) = \frac{CPS \times 100%}{I_\gamma} \times \frac{1}{\text{Activity of the source} \times I_\gamma} \]  

where, \( I_\gamma \) = Absolute gamma ray intensity of the standard source and

\( CPS = \text{Counts per second} = \text{Net peak area per unit time} \)

The energies spread in MCA 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 sources in keV into the MCA to convert all channels to respective energies [11]. The calibration was carried out for a range of gamma-ray energies emitting from the reference source covering the range of interest to make a curve for the efficiency as a function of gamma-ray energy. The empirical equation for the relations between the efficiency (%) and the gamma ray energies are therefore,

\[ Y = 800.1 \times X^{0.96} \]  

(2)

Where, \( Y \) is efficiency expressed in percentage, and \( X \) is gamma-ray energy in keV.

2.3. Counting of Samples and Standards

The software based HPGe detector with digital gamma spectrometry system was used to collect and identify gamma rays and consequently the radioactive species which were produced by them. The counting periods were 20,000 seconds for both samples & standards, and dead time was kept below 5% during counting for analysis. The net count of the sample is obtained by subtracting a linear background distribution of the pulse height spectra from the corresponding peak energy area. From the sample net counts activity of the sample were calculated using the formula,

\[ A = \frac{CPS}{E_{\text{ff}} \times I_r \times W} \]  

(3)

Here, \( A \) is Activity of the sample in Bq/kg, CPS is the net peak counts per seconds, and \( CPS = (CPS \text{ for sample} - CPS \text{ for background value}) \), \( E_{\text{ff}} \) is the peak detection efficiency of the gamma energy, \( I_r \) is absolute intensity of the gamma ray energy and \( W \) is sample’s net weight in Kg. Gamma ray intensities were taken from the literature [11]. The uncertainty of the measurement was premeditated from the counting statistics of the samples. It is done by the formula.

\[ U_w = \frac{Cs \times A}{10^6} \]  

(4)

Where, \( Cs \) is counting statistics and \( Cs = \frac{100 \times \text{CNTs}}{\text{CNTs}} \), CNTs is counting per second, and \( A \) is activity of the sample.

2.4. Annual Effective Dose

The annual effective dose to an individual due to intake of natural radionuclides from the baby food was estimated using the following relationship [12, 1]

\[ D_m = C_m \times CR_m \times D_{cm} \]  

(5)

\( D_m \) is the annual effective dose (Sv·y\(^{-1}\)) due to ingestion of radionuclides from the consumption of baby food, \( C_m \) is the activity concentration of radionuclides in the ingested food (Bq·Kg\(^{-1}\)), \( CR_m \) is the annual intake of milk powder (Kg·y\(^{-1}\)), \( D_{cm} \) is the ingested dose conversion factor (committed effective dose per unit intake via ingestion) for public (Sv·Bq\(^{-1}\)) [1, 13, 14].

3. Results and Discussion

The radioactivity concentration of \(^{238}\text{U}\) and \(^{232}\text{Th}\) were determined via their products under the assumption of secular equilibrium which was reached between \(^{232}\text{Th}\) with its daughters and \(^{238}\text{U}\) & its daughters. The activity concentration of \(^{232}\text{Th}\) was determined from the average activity of \(^{214}\text{Pb}\), \(^{208}\text{Tl}\) and \(^{224}\text{Ac}\), and that of \(^{238}\text{U}\) was determined from the average activity of the \(^{214}\text{Pb}\) and \(^{214}\text{Bi}\) decay products of \(^{228}\text{Ra}\) [15]. Figure 3 shows the \(^{232}\text{Th}\) and
$^{238}$U activity concentration. In milk samples, $^{238}$U concentration varies from 4.4±0.3 to 8.9±0.5 Bq/Kg$^{-1}$ with an average of 5.425±0.4125 Bq/Kg$^{-1}$. In cereals, $^{238}$U concentration varies from 1.7±0.7 to 7.5±0.6 Bq/Kg$^{-1}$ with an average of 2.985±0.38 Bq/Kg$^{-1}$ and $^{232}$Th concentration varies from 2.1±0.56 to 11.3±0.3 Bq/Kg$^{-1}$ with an average 3.94±0.29 Bq/Kg$^{-1}$. Both $^{238}$U and $^{232}$Th activity concentration in milk powder is higher than that in cereals. The radionuclide $^{137}$Cs is one of the tracers to identify the radioactivity in food stuffs including powder milks. For Bangladesh, the maximum permissible limit of radioactivity of $^{137}$Cs in powder milk is less than 94 Bq/kg. In the present experiment, all the samples have been observed to contain $^{137}$Cs activity below the detection limit; these are quite safe for consumption. The findings support radioactivity consequences worldwide.

Due to nuclear accidents like Chernobyl, Three Mile Island, Fukushima, etc., the radionuclides may release in the environment and deposits on the soils. Plants can uptake those radionuclides and enter into the cattle by grazing. Milk from these cattle may contain high radioactivity concentrations. However, in Bangladesh, there is no permissible limit assigned in case of $^{238}$U and $^{232}$Th activity so far. The activity concentration of $^{40}$K, as shown in Figure 4, was the highest in Nido (N.1) powder milk sample, the value being 517.7±16.5 Bq/Kg. The average activity of $^{40}$K in powder milks is almost twice as that of the average in cereals.

The obtained results are compared with other countries (Table 1), the average $^{232}$Th (both in milk and cereals) activity is higher than that of all countries except Saudi Arabia 6.77 BqKg$^{-1}$ [13], whereas $^{238}$U activity for both milk and cereals are very high (Table 1) although most of them exhibit no data about $^{238}$U activity which indicate either a low concentration or not detected due to overlapping of energy spectra with others. In case of $^{40}$K, it is found that both milk and cereals contain high activity like 334.4±12 and 155.7±7.5 BqKg$^{-1}$ respectively, which are lower than that of Brazil, Iran, Nigeria but higher than that of Malaysia, India and almost similar to Israel. Several corrections such as attenuation effect, interference & background effects Uncertainty correction were incorporated for ensuring accuracy of the final results.

In order to assess the Annual effective dose, we need the annual intake by different age group. Almost 100% mothers in Bangladesh breast-fed their infants from beginning to 1 year, every day and different types of milk are added to 60% of infant’s diets by 3 months, and 80% by 6 months of age [16]. The average intake of milk from 1-6 months is about 750 mL per day which is the highest and equivalent to 72 gm of powder milk, the intake of milk decreases the consecutive months depending on taking solid food aside [17, 18].
Table 1. Data table for the international references of natural radionuclides.

<table>
<thead>
<tr>
<th>Country name</th>
<th>40K (Bq/kg)</th>
<th>238U (Bq/kg)</th>
<th>232Th (Bq/kg)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bangladesh (Milk Powder)</td>
<td>334.4±12</td>
<td>5.4±0.4</td>
<td>5.7±0.5</td>
<td>Present Study</td>
</tr>
<tr>
<td>Bangladesh (Cereal)</td>
<td>155.7±7.5</td>
<td>3.0±0.4</td>
<td>4.0±0.3</td>
<td>Present Study</td>
</tr>
<tr>
<td>Malaysia (40.3-254)</td>
<td>99.1</td>
<td>---</td>
<td>2.55 (0.31–8.57)</td>
<td>[19]</td>
</tr>
<tr>
<td>Saudi Arabia</td>
<td>74.5</td>
<td>---</td>
<td>6.77</td>
<td>[13]</td>
</tr>
<tr>
<td>Saudi Arabia</td>
<td>210-257</td>
<td>---</td>
<td>0.096 - 0.76</td>
<td>[20]</td>
</tr>
<tr>
<td>Israel</td>
<td>54.2 - 472.3</td>
<td>---</td>
<td>0.8</td>
<td></td>
</tr>
<tr>
<td>Brazil</td>
<td>475-489</td>
<td>---</td>
<td>1.6-3.6</td>
<td>[21, 22]</td>
</tr>
<tr>
<td>Iran</td>
<td>434.1- 610</td>
<td>0.064</td>
<td>0.094 - 0.166</td>
<td>[23]</td>
</tr>
<tr>
<td>India</td>
<td>34.5±5.2</td>
<td>---</td>
<td>1.48±0.4</td>
<td>[24]</td>
</tr>
<tr>
<td>Nigeria</td>
<td>831.66±54.83</td>
<td>---</td>
<td>4.35±2.06</td>
<td>[25]</td>
</tr>
<tr>
<td>Holland</td>
<td>328.26±4.49</td>
<td>0.65±0.10</td>
<td>1.20±0.20</td>
<td>[23]</td>
</tr>
</tbody>
</table>

The average intake for babies between 12 and 24 months is found to be 14-19 400-550 mL (average 500 mL equivalent to 48 gm) per day [18]. This scenario is for breast feeding and is considered for powder milk since there is no data available for formula milk. Annual effective doses were calculated and compared with the maximum permissible limit in the Table 3 and Figure 5.

Table 2. The Dose Conversion Coefficients for Infant [1].

<table>
<thead>
<tr>
<th>Age group</th>
<th>Effective dose per unit intake Dcm (nSv/Bq)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>238U</td>
</tr>
<tr>
<td>≤1 year</td>
<td>340</td>
</tr>
<tr>
<td>1-2 year</td>
<td>120</td>
</tr>
</tbody>
</table>

Table 3. Annual Effective Dose for Different Age Group.

<table>
<thead>
<tr>
<th>Age group</th>
<th>Sample Type</th>
<th>Annual effective dose (µSv/yr)</th>
<th>Maximum permissible limit [26]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>238U</td>
<td>232Th</td>
<td>40K</td>
</tr>
<tr>
<td>≤1 year</td>
<td>Milk</td>
<td>0.05</td>
<td>0.69</td>
</tr>
<tr>
<td></td>
<td>Cereals</td>
<td>0.02</td>
<td>0.37</td>
</tr>
<tr>
<td>1-2 year</td>
<td>Milk</td>
<td>0.01</td>
<td>0.04</td>
</tr>
<tr>
<td></td>
<td>Cereals</td>
<td>0.01</td>
<td>0.03</td>
</tr>
</tbody>
</table>

Figure 5. Annual Effective Dose (mSv/yr).

Owing to the intake of $^{238}$U, $^{232}$Th and $^{40}$K from milk, the annual effective dose has been estimated as 1.30 mSv/yr$^{-1}$, for the of age group ≤ 1 year which exceed the maximum permissible limit of 1mSv/yr. For cereals the estimated annual effective dose is 0.51 mSv/yr which is below the BAEC recommended value of 1 mSv/yr. For the age group 1-2 year the annual effective dose from Milk and cereals are 0.31 mSv/yr and 0.15 mSv/yr respectively.

4. Conclusion

The natural radioactivity in powder milk and cereals samples and annual effective dose corresponding to two different infants groups based on their age have been
estimated by the gamma spectroscopy. It has been observed that annual effective dose achieve from milk and cereals consumption, are for age group ≤ 1 year is 1.30 mSv/yr and 0.51 mSv/yr and for age group 1-2 year 0.31 mSv/yr and 0.15 mSv/yr respectively. The dose received from milk for ≤1 year is higher than the WHO recommended limit of 1 mSv.y⁻¹. The data generated in this study will provide base-line radiometric values of activity concentration and annual effective dose attributed from baby foods in Bangladesh and further study is required for precise data and infant’s health safety.

References


