Assessment of the Activity concentrations of the naturally occurring radionuclides from the samples collected from the different (CUFL & JFL) Urea Fertilizer Company Limited

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ABSTRACT

The activity concentrations of ^{238}U, ^{232}Th and ^{40}K have been determined for product (urea) samples collected from the Chittagong Urea Fertilizer Limited (CUFL) and Jamuna Fertilizer Company Limited. The different kinds of sample were analyzed by using a calibrated high purity germanium (HPGe) detector of relative efficiency of 38%. For solid samples, the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K have been found 78.67±2.87, 59.74±2.27 and 463.71±9.16 Bq/Kg, respectively. The mean activity concentrations of product samples for the corresponding radionuclide were 73.12±2.83, 74.59±2.32 and 391.58±9.38 Bq/Kg, respectively. The artificial radionuclide ^{137}Cs has not been detected in any of the samples. The obtained results of this study show that for some samples the values are lower and for the remaining are higher than the World average values. The mean representative index value is less than unity which confirms that the area is safe for the factory workers and the public nearby.

Keywords: Activity Concentration, Natural Radionuclides, Gamma Ray Spectroscopy, World Average Values

INTRODUCTION

The uranium and thorium series, and 40K are mainly natural radionuclides in soil, building materials and recycled industrial waste (Gibson, Thompson, & Spiers, 1993; report, 1988). It is important to calculate radioactive parameters and contents in the construction materials due to public exposure. Natural radionuclide in soil contributes a significant amount of background radiation exposure to the population through inhalation and ingestion. The main contributors of radionuclides are ^{238}U, ^{232}Th and ^{40}K. Since these radionuclides in soils are not uniformly distributed and vary from region to region (Gibson et al., 1993; report, 1988). Natural radioactivity is a part of our nature surrounding us and concentrations of natural radionuclides in the environment increase with the development of technologies. Fertilizer industries, the phosphate fertilizer industries, are important sources of exposure to ionizing radiation of the people and possible contamination of the environment. Therefore, the objective of the present study is to detect the radionuclides and determine their concentration levels present in the wastes generated from the Chittagong Urea Fertilizer Limited and Jamuna Fertilizer Company Limited on the workers of the factory and the public nearby and the associated environment.

METHODS AND MATERIALS

Study Area
The samples were collected from Chittagong Urea Fertilizer Limited and Jamuna Fertilizer Company Limited. Karnofully river flows by the side of the CUFL factory and Jamuna river flows by the side of the Jamuna Fertilizer Company Limited, it is the main source of local irrigation. The location of this factory is shown in Fig. 1.1 & Fig.1.2.

Fig 1.1 Geographical position & Factory of Chittagong Urea Fertilizer Limited (CUFL)

Fig 1.2 Factory of Jamuna Fertilizer Company Limited

SAMPLE COLLECTION AND PREPARATION

The different types of sample, namely; clay (clarifier out, S-1 & S-2), first reformer catalyst(Ni, S-3), and final product (Urea, P-1 & P-2), we were recollected from Chittagong Urea Fertilizer Limited (CUFL) factory. Two other product urea samples were collected from Jamuna Fertilizer Company Limited (P-3 & P-4). Standard methods were followed to process the samples for characterization (IAEA Technical Report No. 295, 1989). The soil samples were dried in the sun for several days. The samples were then crushed and dried again in a temperature controlled oven at 100^0C for 24 hours in order to remove the moisture content in the samples. The dried samples were ground to fine powder and passed through a sieve of mesh size 200μm. The samples were then filled in cylindrical plastic containers of 6 cm diameter and 7 cm height with a volume of 180 ml as shown in Fig. 2.The weights of all the samples were taken by an electronic balance and the net weights of the samples were noted as before. Finally, the plastic containers were closed by caps and wrapped with thick vinyl tape about their necks to seal the containers tightly. The samples were then stored for about 30 days to assure secular equilibrium between the ^{238}U and ^{232}Th series and their daughters progeny (Tufail, Iqbal, & Mirza, 2000).

Fig 2. Solid samples.

GAMMA-RAY DETECTION SYSTEM

In γ-ray spectrometry, the full energy peak efficiency of a high purity Germanium (HpGe) detector is the number of γ- rays detected by the detector to the number of photons emitted by the source for a specific energy, which is defined as (Kannan, Rajan, Iyengar, & Ramesh, 2002)

$$\in (E) = \frac{n(E)}{A \times I_\gamma} \qquad (1)$$

where, n(E) is the net count rate of the photo peak for the corresponding energy E , A is the present activities of the standard reference source which were calculated by using the well-known decay law: A=A_0 $e^{-\lambda t}$. and I_γ is the Intensity of the gamma energy. For the determination of efficiency of the detector (HpGe), the contributions for the coincidence summing effect and the angular correlations due to the cascading gamma-rays were taken into account followed by the literatures (Siegbahn, 2012). In the present study, the International Atomic Energy Agency (IAEA) reference samples were used for the calibration of detector efficiencies. The IAEA reference samples are: RGU-1, Uranium is in silica matrix, RGTh-1: Thorium is in silica matrix and RGK-1: potassium sulphate. The standard reference source has the same diameter as the soil samples of known concentrations of ^{238}U, ^{232}Th and ^{40}K radionuclides supplied by the Canada Centre for Mineral and Energy Technology (CAMET) under a contract with the IAEA.

SAMPLE ANALYSIS

The detection and measurement of radionuclides in the samples were carried out by gamma spectrometry system using a vertical coaxial cylindrical HPGe detector of 172 cm3 active volume with 38% relative efficiency. The p-type HPGe detector supplied by CANBERRA (Model IGG 2020) had a resolution of 2 keV at 1332 keV of ^{60}Co. The detector was coupled to a 16 k-channel computer analyzer. The analysis was

carried out using a Genie 2000 software, which matched various gamma energy peaks to a library of possible radionuclides. The detector was enclosed in a cylindrical shielding container made of lead and iron with 11.3 cm thickness, 51 cm height and 28 cm internal diameter and with a fixed bottom and moving cover to reduce the external γ-ray background (Islam *et al,*. 1990). All the samples were counted for 50 ks. Prior to the measurement of the samples, the gamma background at laboratory site was determined with an identical empty Marinelli beaker and plastic container used in the sample measurement. The energy regions selected for the corresponding radionuclides were 295 keV and 352 keV of ^{214}Pb and 609 keV, 1120 keV and 1764 keV of ^{214}Bi for ^{226}Ra, 583 keV and 2614 keV of ^{208}Tl, 911 keV and 969 keV of ^{228}Ac for ^{228}Th and 1460 keV for ^{40}K (Roessier *et al,*. 1970).

CALIBRATION OF HPGE DETECTOR

In the present study the calibration for the efficiency of the detector was performed by the International Atomic Energy Agency (IAEA) reference samples of solid and liquid. The IAEA reference samples are: RGU-1, Uranium is in silica matrix, RGTh-1: Thorium is in silica matrix and RGK-1: potassium sulphate.
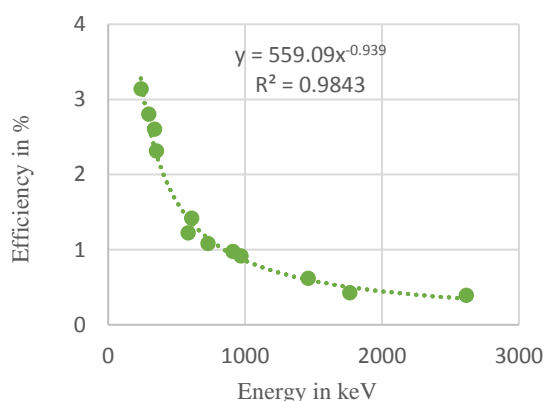

$y = 559.09x^{-0.939}$
$R^2 = 0.9843$

Fig 3.Counting efficiency curve of the HPGe Detector

The standard reference source has the same diameter as the soil samples of known concentrations of ^{238}U, ^{232}Th and ^{40}K radionuclides supplied by the Canada Centre for Mineral and

Energy Technology (CAMET) under a contract with the IAEA. The detector efficiency calibration curves as a function of energy for both liquid and solid matrices have been shown in Fig. 3.

CALCULATION OF ACTIVITY CONCENTRATION

The radioactivity of each sample was measured using the calibrated high purity Germanium (HpGe) Detector of energy resolution of 2.0 KeV at 1.33 MeV of ^{60}Co. for a period of 20,000 s. Keeping the samples one by one on the top of the detector and counted for a period of 20,000 s. The activity concentration (A) of each radionuclide in the sample was determined by using the net count rates (Nc) after subtracting the background counts from the gross counts 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**,**

$$A = (Nc \times 1000) / (\varepsilon \times I\gamma \times W) \qquad (2)$$

Where, A = Activity concentration of the sample in Bq/.Kg Net count rate, Nc = Gross counts per second from the samples - background counts per second ε = Efficiency of the detector for the specific energy. $I\gamma$ = Intensity of the gamma ray. W = Sample weight in gm. For the analysis of peak areas of gamma spectra, a Computer software programming (GENIE 2000) was used.

RESULTS AND DISCUSSION

Activity Concentrations of all Samples

Activity concentrations for nuclides ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in Chittagong Urea Fertilizer limited (CUFL) samples were determined by equation (2) and the results for the same have been shown in Table 1 with the uncertainty of 1σ level. Here, the table shows that the highest value is found for sample S-3. The highest value of the nuclide may vary from place to place and this variation may it is a chemical composite (First Reformer Catalyst ,Ni). Moreover, it is also possible due to the excess use of agricultural fertilizers and pesticides. The results for the nuclides (^{238}U, ^{232}Th and ^{40}K) are also shown independently through Figure 3.

Table-1: Activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs of type of samples.

Sample Type	Sample ID	Activity of ^{238}U, (Bq.Kg^{-1}) with ($\pm 1\sigma$)	Activity of ^{232}Th, (Bq.Kg^{-1}) with($\pm 1\sigma$)	Activity of ^{40}K, (Bq.Kg^{-1}) with ($\pm 1\sigma$)	Activity of ^{137}Cs, (Bq.Kg^{-1}) with ($\pm 1\sigma$)
Solid	S-1	77.04±2.84	61.85 ±2.16	520.85±10.4	ND
	S-2	72.04±2.81	52.16 ±2.12	410.96±9.41	ND
	S-3	86.94±2.91	65.21 ±2.44	459.31±7.68	ND
	Mean	78.67±2.87	59.74±2.27	463.71±9.16	ND
Product urea (CUFL)	P-1	72.09±2.82	73.14 ±2.25	387.90±11.3	ND
	P-2	74.12±2.83	76.03 ±2.36	395.58±7.45	ND
	Mean	73.12±2.83	74.59±2.32	391.58±9.38	ND
Product urea (Jamuna)	P-3	74.31±2.82	57.60 ±1.93	388.99±11.4	ND
	P-4	69.91±2.79	57.31 ±1.89	317.56±9.04	ND
	Mean	72.11±2.81	57.46±1.91	353.28±10.22	ND

^{238}U Activity

In all samples, the activity concentrations of ^{238}U were found in the range of 17.86±0.79− 86.94±2.91Bq/kg, (where, for solid samples 72.04±2.81−86.94±2.91 Bq/Kg, for product samples 69.91±2.79−74.31±2.82 Bq/Kg, for liquid samples 17.86±0.79−28.91±1.97 Bq/Kg) with mean value of 41.86± 1.82 Bq/kg . This result is also higher than the world average of 35 Bq/kg for the same radionuclide in soils reported by UNSCEAR (Unscear, 2000).
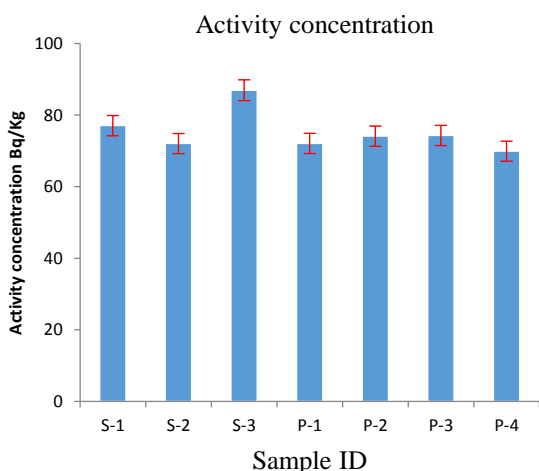

Fig 4. Activity concentration of all samples with world average value.

^{232}TH ACTIVITY

The activity concentrations of ^{232}Th is found in the range of 7.33 ±1.89− 76.03 ±2.36 Bq/kg (where, for solid samples 52.16 ±2.12−65.21 ±2.44 Bq/Kg, for product samples 57.31 ±1.89−76.03 ±2.36 Bq/Kg, for liquid samples 7.33 ±1.89−13.41 ±1.89 Bq/Kg) with mean value of **33.12± 1.94** Bq/kg . This result is also higher than the world average of 30 Bq/kg (Siegbahn, 2012).
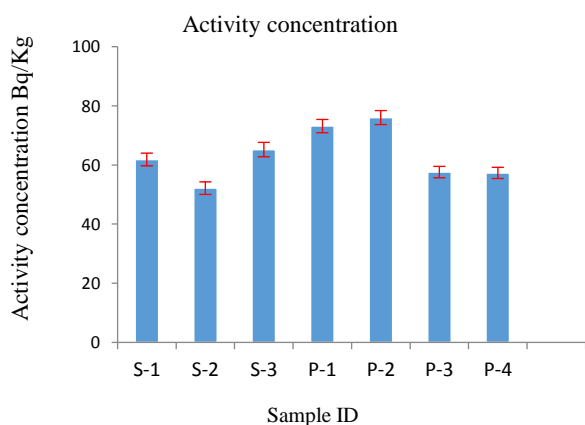

Fig 5: Activity concentration of all samples with world average value.

^{40}K ACTIVITY

The activity concentrations of ^{40}K is found in the range of 19.07±1.36-520.85±10.4 Bq/kg (where, for solid samples 410.96±9.41−520.85±10.4 Bq/Kg, for product samples 317.56±9.04−395.58±7.45 Bq/Kg, for liquid samples 19.07±1.36−63.32±1.95 Bq/Kg) with mean value of 193.32±7.84 Bq/kg . This result is slightly lower than the worldwide average of 400Bq/kg for the same kind of nuclide (Karahan & Bayulken, 2000).
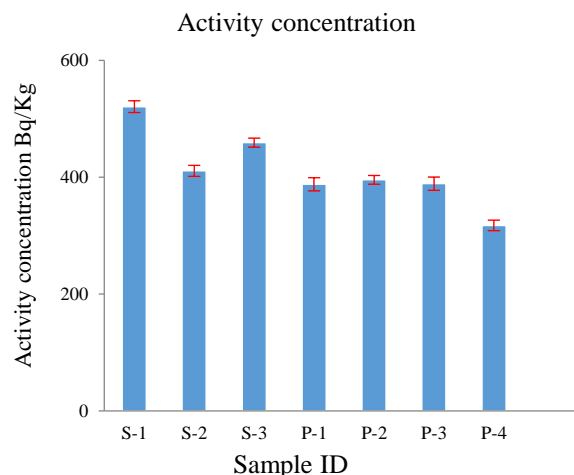

Fig 6. Activity concentration of all samples with world average value.

^{137}CS ACTIVITY

The activity concentration of ^{137}Cs is not found in any types of the samples.
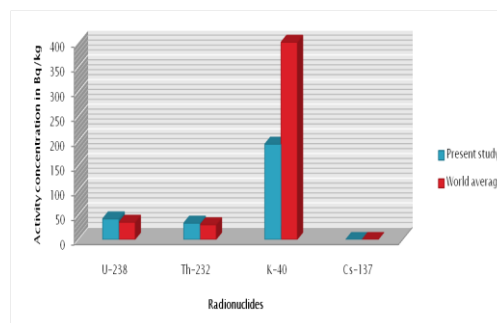

Fig 7. Mean activity concentration of all samples with world average value.

CONCLUSION

The detection of radionuclides, their activity concentrations of different kinds of (solid and product) samples from Chittagong Urea Fertilizer Limited and Jamuna Fertilizer Company Limited from the local market were determined. The detection of natural radionuclides of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs and their activity concentrations were determined by using a calibrated HpGe detector. Here there is no artificial radionuclide was found in this study. The activity concentrations for all samples were determined individually. The activity concentration values for a few samples were lower and for others were higher than the world average values.

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