



## An Environmental Impact Study of Jamuna Urea Fertilizer Factory at Tarakandi in Jamalpur with Radiological Indices

M. A. Samad<sup>1</sup>, M. I. Ali<sup>2</sup>, D. Paul<sup>2</sup> and S. M. A. Islam<sup>1</sup>

<sup>1</sup>Department of Physics, Jahangirnagar University, Savar, Dhaka

<sup>2</sup>Health Physics and Radioactive Waste Management Unit, Institute of Nuclear Science and Technology, Atomic Energy Research Establishment, Savar, Dhaka

### Abstract

The activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K have been determined in solid and liquid samples collected from the Jamuna Urea Fertilizer Factory, Tarakandi, Jamalpur, Bangladesh. Six different types of samples, namely, i) the liquid waste water stored inside the factory ii) water sample near the liquid waste disposal point of Jamuna river iii) liquid-waste-mixed soil iv) normal soil adjacent to the factory v) the final product (Urea) and vi) urea dust of the factory have been analyzed by gamma spectrometric technique using a high purity germanium (HPGe) detector of 40% relative efficiency. The activity concentration of <sup>226</sup>Ra was determined from the average concentrations of the <sup>214</sup>Pb and <sup>214</sup>Bi decay products, the activity concentration of <sup>232</sup>Th was determined from the average concentrations of <sup>208</sup>Tl and <sup>228</sup>Ac, whereas the concentration of <sup>40</sup>K was measured directly. The analysis of the liquid waste samples showed that the average activity concentrations of <sup>226</sup>Ra and <sup>232</sup>Th were  $3.64 \pm 0.72$  and  $12.94 \pm 2.02$  Bq/L, respectively and no <sup>40</sup>K was detected in any of the samples. In the liquid-waste-mixed soil samples the average value of activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were  $21.45 \pm 3.19$ ,  $63.00 \pm 8.10$  and  $311.97 \pm 90.93$  Bq/kg, respectively whereas in the normal soil the values were  $20.97 \pm 3.56$ ,  $61.77 \pm 8.93$  and  $645.48 \pm 103.62$  Bq/kg, respectively. Considering the stored wastes as a part of the ambient environment of the factory, radium equivalent activity, radiation hazard index and external annual effective dose to the worker and the public due to these wastes were calculated and compared with the world average values.

**Key words:** Activity concentration, Environment, Detector, Radium equivalent, Dose

### Introduction

A number of human practices and non-nuclear industries contribute to further concentration of some of the natural radionuclides that can be found in the earth's crust affecting the human and the environment. 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, specifically the phosphate fertilizer industries, are important sources of exposure to ionizing radiation of the people and possible contamination of the environment. The production process of fertilizer redistributes radionuclides throughout the environment and introduces them into the final products and byproducts (Saueia *et al.*, 2005). Therefore, it is worthy of investigating the radionuclide concentration in the final products and byproducts of the fertilizer and assess the radiological impact of fertilizer industries on the workers, public and the environment.

Urea fertilizer industries form an important segment of total chemical industry in Bangladesh (Alam *et al.*, 1997). Urea fertilizers are used of a huge amounts in Bangladesh in order to grow more food to meet the increasing demand of food. There are seven urea fertilizer factories scattered all over Bangladesh involving vast area and huge manpower. Jamuna Urea Fertilizer Factory, Tarakandi, Jamalpur, is an important installation in the country contributing significantly to the production of urea.

Jamuna Urea Fertilizer Factory uses natural gas as the starting material along with some catalysts such as Fe<sub>2</sub>O<sub>3</sub>, Fe<sub>3</sub>O<sub>4</sub>, Al<sub>2</sub>O<sub>3</sub>, K<sub>2</sub>O, CaO and SiO<sub>2</sub> to produce urea and these are the parts of the naturally occurring radioactive materials (NORMs) inevitably present in nature from the formation of earth (EFMA 2000, <http://www.teacher.buet.ac.bd.>, Ismail *et al.*, 2009). Moreover, the liquid wastes generated from the production process are discharged from the complex to the environment which finally goes to the Jamuna River and to the cultivated land adjacent to the complex. Contamination of land and water can occur either from deposition of material originally introduced into the atmosphere or from waste products discharged directly to the ground, surface or subsurface waters from which they are eventually mobilized by ground water or erosion. Thus the wastes generated from fertilizer factories may contaminate our environment and eventually causes radiation exposure to the public. There are some reports on the presence of radioactivity in the chemical fertilizer used in Bangladesh but corresponding data on radiation exposure due to these fertilizer and the wastes generated from the fertilizer industries are still very less (Henshaw 1990).

The analyses of radionuclide concentrations and activity level would provide us some important information to evaluate the extent, degree and the different pathways for the radioactive contamination to interrupt the natural system. Therefore, the objective of the present study is to detect the radionuclides and determine their concentration levels

present in the wastes generated from the Jamuna Urea Fertilizer Factory, Tarakandi, Jamalpur and to estimate their radiological impact on the public and the associated environment.

## Materials and Methods

### Sampling sites

The samples were collected from Jamuna Urea Fertilizer Factory, Tarakandi, Jamalpur. The factory is situated at the northern side of Bangladesh. The geographical location of the study area as determined by a hand held GPS set are: latitude 24°41' N and longitude 89°50' E. The factory is surrounded by tea gardens and farmlands. Jamuna River flows by the side of the factory and it is the main source of local irrigation.

### Sample collection and preparation

Six different types of samples, namely; liquid waste, river water (waste mixed and fresh), liquid-waste-mixed soil, normal soil adjacent to the factory, urea dust and final product (Urea), were while the total number of samples we recollected from Jamuna Urea fertilizer factory, Tarakandi, Jamalpur. Standard methods were followed to process the samples for characterization (IAEA Technical Report No. 295, 1989) Twenty.

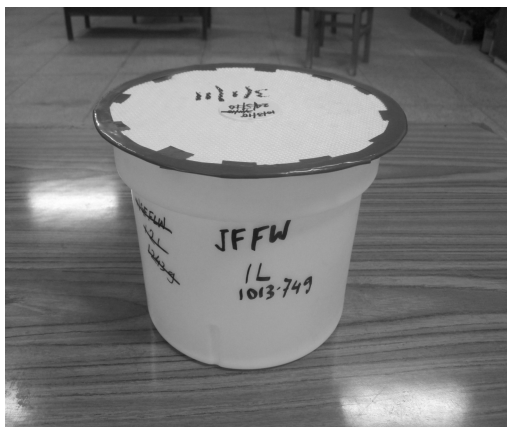
### For liquid samples

Marinelli type beakers (2 liter capacity) were used to process and measure the liquid samples as shown in Fig. 2(a). At the start of the sample processing steps,

the beakers were made contamination-free by light hydrochloric acid solution and deionized water. The beakers were then dried using a temperature-controlled oven and the weights of the empty beakers were noted. The beakers were then filled with liquid samples. The net weights of the samples were found from the difference of weights of sample-filled and empty beakers. The beakers filled with samples were then kept in the sun for several days. Finally, the beakers were closed by caps, sealed tightly and wrapped with thick vinyl tape and kept for 30 days for achieving the secular equilibrium between gaseous and non-gaseous decay products of naturally occurring radioactive series.

### For solid samples

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°C 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 200 µm. 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(b). 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  $^{238}\text{U}$  and  $^{232}\text{Th}$  series and their daughters progeny (Tufail *et al.*, 2000).



(a)



(b)

Figure 2. Photographs of the sample containers (a) liquid and (b) solid samples

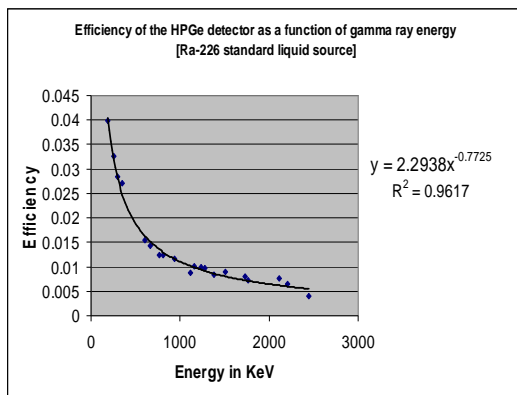
**Sample analysis**

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 cm<sup>3</sup> active volume with 40% relative efficiency. The p-type HPGe detector supplied by CANBERRA (Model GC4020) had a resolution of 2 keV at 1332 keV of <sup>60</sup>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  $\gamma$ -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

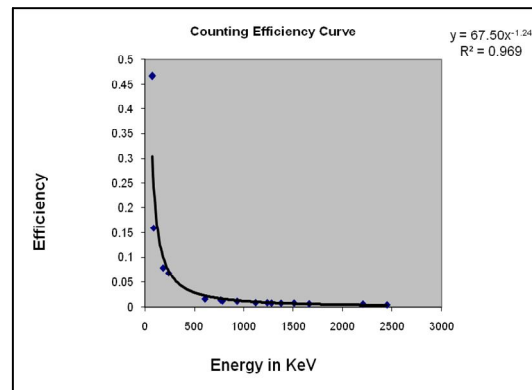
selected for the corresponding radionuclides were 295 keV and 352 keV of <sup>214</sup>Pb and 609 keV, 1120 keV and 1764 keV of <sup>214</sup>Bi for <sup>226</sup>Ra, 583 keV and 2614 keV of <sup>208</sup>Tl, 911 keV and 969 keV of <sup>228</sup>Ac for <sup>228</sup>Th and 1460 keV for <sup>40</sup>K (Roessier *et al.*, 1970).

**Calibration of HPGe detector**

In the present study the calibration for the efficiency of the detector was performed by standard sources of solid and liquid matrices prepared using <sup>226</sup>Ra standard solutions (Tahawy *et al.*, 1992). The standard sources were prepared using identical containers used for the measurement of the samples, e.g., 2L Marinelli beakers for water and plastic container for soil. The preparation process of standard sources has been reported elsewhere (Ahmed 2010). The detector efficiency calibration curves as a function of energy for both liquid and solid matrices have been shown in Fig. 3. The energy calibration of the detector was performed by <sup>137</sup>Cs and <sup>60</sup>Co point sources.



(a)



(b)

**Figure 3 Efficiency calibration curve of HPGe detector for (a) liquid and (b) solid matrices**

**Calculation of activity concentration**

The net amount of a sample had been 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 samples were calculated using the formula (Usif and Taher 2008):

$$A = \frac{cps}{E \times I \times w} \text{----- (1)}$$

Where, A = Activity of the sample in Bq.kg<sup>-1</sup> or Bq.L<sup>-1</sup>

cps = the net counts per second

= cps for the sample- cps for background value

E = the counting efficiency of the gamma energy

I = absolute intensity of the gamma ray and

w = samples net weight (in kg)

The errors in the measurement have been expressed in terms of standard deviation ( $\pm \sigma$ ), where  $\sigma$  is expressed as (Knoll 1998):

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

Where,  $N_s$  is the counts measured in time  $T_s$  and  $N_b$  is the background counts measured in time  $T_b$ . The standard deviation  $\pm \sigma$  in cps was converted into activity in Bq.kg<sup>-1</sup> according to equation (1).

### Results and discussions

The radionuclides detected and corresponding activity concentrations in different samples, namely, liquid waste, Jamuna river normal water and water waste mixed point, liquid-waste-mixed soil, normal soil, final product (Urea) and urea dust have been summarized in **Table 1** to **Table 6**, respectively.

The radionuclides found in the samples were  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ ,  $^{228}\text{Ac}$ ,  $^{208}\text{Tl}$  (due to  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series) and  $^{40}\text{K}$ . These are only natural radionuclides present in the samples and no artificial radionuclide, such as  $^{137}\text{Cs}$ , was found in any of the samples.

**Table 1** shows the activity concentrations of the radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  detected in the four liquid waste samples. The average values of activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  in these samples were found to be  $3.64 \pm 0.72$  and  $12.94 \pm 2.02$ , respectively. It was also observed that no  $^{40}\text{K}$  was found in any of the four samples. The activity concentrations of the water collected from the Jamuna river have been shown in **Table 2**. Two types of water samples were

collected from the river, namely, the water sample from the liquid waste disposal point and normal water point. The average concentrations of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the samples were found to be  $1.83 \pm 0.56$ ,  $8.64 \pm 1.53$ , respectively in the samples collected from the waste disposal point and no  $^{40}\text{K}$  was found in any of the two samples collected from this point. The activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the samples collected from the normal water of Jamuna river were  $1.59 \pm 0.52$ ,  $1.76 \pm 1.42$  and  $11.34 \pm 4.73$ , respectively. The results on the liquid samples (liquid waste, waste-mixed river water and normal river water) revealed that there was no  $^{40}\text{K}$  in the liquid waste or waste-mixed river water but it was found in the normal river water. The results also showed that the concentration levels of the radionuclides between the liquid waste sample and water samples at the waste disposal point were almost the same. However, the concentration levels of the radionuclides in the samples collected from the normal water were slightly different.

**Table 1. Activity concentrations of the radionuclides in liquid waste samples collected from the factory**

Samples ID	Activity concentrations of radionuclides (Bq/L)		
	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{40}\text{K}$
Sample:1	$2.48 \pm 0.50$	$6.79 \pm 1.41$	ND
Sample: 2	$5.39 \pm 0.80$	$12.14 \pm 2.14$	ND
Sample: 3	$5.12 \pm 0.79$	$12.53 \pm 2.15$	ND
Sample: 4	$1.58 \pm 0.76$	$16.56 \pm 2.19$	ND
Sample: 5	$3.64 \pm 0.74$	$16.66 \pm 2.19$	ND
<b>Average</b>	<b><math>3.64 \pm 0.72</math></b>	<b><math>12.94 \pm 2.02</math></b>	<b>ND</b>

ND: Not Detected.

**Table 2. Activity concentrations of radionuclides in Jamuna river water**

	Samples ID	Activity concentrations of radionuclides (Bq/L)		
		$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{40}\text{K}$
Waste disposal point	Sample: 6	$1.12 \pm 0.55$	$9.02 \pm 1.53$	ND
	Sample: 7	$2.53 \pm 0.56$	$8.25 \pm 1.52$	ND
	<b>Average</b>	<b><math>1.83 \pm 0.56</math></b>	<b><math>8.64 \pm 1.53</math></b>	<b>ND</b>
Normal water point	Sample: 8	$1.56 \pm 0.52$	$1.93 \pm 1.43$	$8.09 \pm 2.21$
	Sample: 9	$1.61 \pm 0.52$	$1.59 \pm 1.41$	$14.58 \pm 7.24$
	<b>Average</b>	<b><math>1.59 \pm 0.52</math></b>	<b><math>1.76 \pm 1.42</math></b>	<b><math>11.34 \pm 4.73</math></b>

ND: Not Detected

The activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the liquid-waste-mixed soil samples have been shown in **Table 3**, whereas the activity concentrations of those in the normal soil samples collected from the adjacent area of the factory have been shown in **Table 4**. The average concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the liquid-waste-mixed soil samples were found to be  $21.45 \pm 3.19$ ,  $63.00 \pm 8.10$  and  $311.97 \pm 90.93$ , respectively whereas in the normal soil samples were

$20.97 \pm 3.56$ ,  $61.77 \pm 8.93$  and  $645.48 \pm 103.62$ , respectively. The comparison between these two shows that higher average concentration levels of  $^{232}\text{Th}$  and  $^{40}\text{K}$  and lower average concentration level of  $^{226}\text{Ra}$  were present in the waste-mixed soil sample than that of normal soil samples. However, the differences in the concentration levels of the radionuclides between these two types of samples were not much significant.

**Table 3. Activity concentrations of radionuclides in liquid-waste-mixed soil samples collected from the different locations of the factory**

Samples ID	Activity concentrations of radionuclides (Bq/kg)		
	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
Sample: 10	11.96 ± 3.12	18.83 ± 7.65	210.77 ± 91.14
Sample: 11	21.96 ± 3.22	66.96 ± 8.01	327.87 ± 91.64
Sample: 12	30.44 ± 3.23	103.22 ± 8.65	397.26 ± 90.00
<b>Average</b>	<b>21.45 ± 3.19</b>	<b>63.00 ± 8.10</b>	<b>311.97 ± 90.93</b>

**Table 4. Activity concentrations of radionuclides in normal soil samples collected from different locations adjacent to the factory**

Samples ID	Activity concentrations of radionuclides (Bq/kg)		
	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
Sample: 13	11.69 ± 3.47	31.06 ± 8.66	499.03 ± 100.88
Sample: 14	29.39 ± 3.69	79.10 ± 9.11	758.39 ± 105.15
Sample: 15	21.84 ± 3.53	75.15 ± 9.02	679.02 ± 104.82
<b>Average</b>	<b>20.97 ± 3.56</b>	<b>61.77 ± 8.93</b>	<b>645.48 ± 103.62</b>

**Table 5. Activity concentrations of the radionuclides in the final product (Urea)**

Samples ID	Activity concentrations of radionuclides (Bq/kg)		
	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
Sample:16	15.47 ± 2.99	12.49 ± 6.61	ND
Sample:17	16.50 ± 3.00	18.17 ± 6.67	ND
Sample: 18	14.17 ± 2.83	20.81 ± 6.90	ND
<b>Average</b>	<b>15.38 ± 2.94</b>	<b>17.16 ± 6.73</b>	<b>ND</b>

**Table 6. Activity concentrations of the radionuclides in the Urea dust sample**

Samples ID	Activity concentrations of radionuclides (Bq/kg)		
	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
Sample:19	14.17 ± 3.32	20.81 ± 6.80	ND
Sample: 20	15.22 ± 3.04	28.90 ± 6.87	ND
<b>Average</b>	<b>14.69 ± 3.33</b>	<b>24.86 ± 6.84</b>	<b>ND</b>

ND: Not Detected

The activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in urea and urea dust samples collected from the factory have been summarized in **Table 5 and Table 6**. The values for <sup>226</sup>Ra and <sup>232</sup>Th found in the present study were slightly higher than the other report (Alam *et al.*, 1997). However, no <sup>40</sup>K radionuclide had been detected in the urea samples in the present study. Urea is produced from natural gas, nitrogen and water along with some catalysts as stated earlier, the presence of small amount of these radionuclides may be due to the impurities present in the raw materials.

The wastes (liquid waste and liquid-waste-mixed soil) generated from the production process of the urea fertilizer had been stored in the open environment;

therefore, these wastes with natural radionuclides had become a constituent of the ambient environment of the factory complex. Therefore, these wastes could be treated as the part of environment contributing to the exposure to the workers of the factory and to the general public. Therefore, the radium equivalent activity ( $Ra_{eq}$ ), external radiation hazard index ( $H_{ex}$ ), absorbed gamma dose rate (D) and annual effective dose (E) were calculated for the studied samples, by using the following equations below (Kessaratikoon and Awaekchi 2008) and the results have been presented in **Table 6**.

$$Ra_{eq} \text{ (Bq/L)} = A_{Ra} + 1.43A_{Th} + 0.07A_K \dots\dots\dots(3)$$

$$H_{ex} = A_{Ra} / 370 + A_{Th} / 259 + A_K / 4810 \dots\dots\dots(4)$$

$$D \text{ (nGy/h)} = 0.462A_{Ra} + 0.604A_{Th} + 0.042A_K \dots\dots(5)$$

$E (Sv) = D \times 24 \times 365 \times 0.7 \times 0.2 \dots\dots\dots(6)$

It was seen from the results that the values of  $R_{a_{eq}}$ ,  $H_{ex}$ , D and E were lower for the case of liquid waste

samples than those of solid samples. The values of these parameters for both liquid-waste-mixed-soil samples and normal soil samples were almost same.

**Table 7. Radium equivalent activity, external radiation hazard index, absorbed dose rate and annual effective dose for liquid waste, liquid-waste-mixed-soil and normal soil**

	Sample ID	$R_{a_{eq}}$ (Bq/L)	$H_{ex}$	D (nGy/h)	E (mSv/y)
<b>Liquid waste</b>	Sample: 1	12.19	0.033	5.25	0.006
	Sample: 2	22.75	0.062	9.82	0.012
	Sample: 3	23.04	0.062	9.93	0.012
	Sample: 4	25.26	0.068	10.73	0.013
	Sample: 5	27.46	0.074	11.74	0.014
	<b>Average</b>	<b>22.14</b>	<b>0.060</b>	<b>9.49</b>	<b>0.011</b>
	Sample ID	$R_{a_{eq}}$ (Bq/Kg)	$H_{ex}$	D (nGy/h)	E (mSv/y)
<b>Liquid-waste-mixed soil</b>	Sample: 10	53.62	0.149	25.75	0.032
	Sample: 11	140.66	0.386	64.36	0.078
	Sample: 12	205.85	0.563	93.09	0.114
	<b>Average</b>	<b>133.38</b>	<b>0.366</b>	<b>61.07</b>	<b>0.075</b>
<b>Normal soil</b>	Sample: 13	91.03	0.255	45.12	0.055
	Sample: 14	195.58	0.542	93.20	0.113
	Sample: 15	176.82	0.490	83.99	0.103
	<b>Average</b>	<b>154.48</b>	<b>0.429</b>	<b>74.10</b>	<b>0.091</b>

The world average values for  $R_{a_{eq}}$ ,  $H_{ex}$ , D and E are 370 Bq/Kg, 1.0, 55 nGy/h and 0.460 mSv/y, respectively for environmental samples (ICRP, 1990). In the present study, the maximum value of annual effective dose was 0.114 mSv/y and it was found in the liquid waste mixed soil sample. This value of annual effective dose rate is much below the standard value set by ICRP for the general public (ICRP, 1990). Therefore, the study revealed that the factory complex

of Jamuna Urea Fertilizer Factory and its adjacent areas are within normal natural background radiation area and no excess radionuclide concentration was found in the samples collected from this factory. This study could be useful as a baseline data for the radiological impact of the wastes generated from the urea fertilizer industries of Bangladesh on the members of public and the environment.

**Conclusion**

The detection of radionuclides and their activity concentrations in six different types of samples collected from Jamuna Urea Fertilizer Factory, Tarakandi, Jamalpur, had been determined as a part of assessment of the radiological impact of fertilizer industries of Bangladesh on the public and the environment. The radionuclides detected in the samples were all natural and no artificial radionuclide was found in the study. The activity concentrations of the detected radionuclides and the doses from both solid and liquid waste samples were found comparable to the normal samples. However, the values found in the present study were lower than the

world average values. The annual effective doses from these samples were found within the acceptable limit set by ICRP. This study could be useful as a baseline data for radionuclide concentration and radiation exposure to fertilizer wastes generated from the fertilizer industries of Bangladesh.

**Acknowledgement**

The authors would like to thank the Health Physics and Radioactive Waste Management Unit, AERE, Savar, Dhaka for allowing us to use their laboratory facility and the University Grants Commission, Bangladesh for the financial assistance to conduct the research.

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