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Article

Assessment of radioactive pollution around a fertilizer factory complex in the North-Eastern part of Bangladesh

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ABSTRACT **The activity concentrations of soil, water and fertilizer samples were determined by using high-resolution gamma spectrometry (HPGe detector: 40% relative efficiency) with a PC-based MCA system. The samples were collected from the area of a urea fertilizer factory, lagoon and Shitalakhya river in Narsingdi, Bangladesh. The activity concentrations of ^{226}Ra ranged from 3.16 ± 0.32 to 10.28 ± 0.55 Bq.kg^{-1}, 1.22 ± 0.41 to 7.36 ± 0.42 Bq.L^{-1} and 3.55 ± 0.33 to 90.65 ± 3.17 Bq.kg^{-1} for soil, water and fertilizer samples, respectively. The ^{232}Th activity concentrations ranged from 4.89 ± 0.45 to 15.82 ± 0.45 Bq.kg^{-1}, 1.21 ± 0.06 to 8.59 ± 0.37 Bq. L^{-1} and 4.76 ± 0.25 to 26.38 ± 1.40 Bq.kg^{-1} for soil, water and fertilizer samples, respectively. The ^{40}K activity concentrations ranged from 24.96 ± 0.23 to 60.49 ± 0.56 Bq.kg^{-1}, 7.48 ± 0.53 to 35.48 ± 0.24 Bq.L^{-1} and 3.55 ± 0.05 to 3051.71 ± 19.53 Bq.kg^{-1} for these samples, respectively. The radium equivalent activity, the hazard indices, the gamma activity concentration index, the indoor absorbed dose rate and the corresponding annual effective dose were estimated for the potential radiological hazard of the collected samples. The calculated values of the representative level index values (Iγr) for all samples of the study area are lower than unity except the MOP sample. The activity ratios were also measured. These values are compared with reported values for other countries of the world. The results of the comparison studies show that the radioactivity concentrations and other radioactive indices of the samples of the study area are below the internationally accepted maximum permissible values. Therefore, this region is safe from any radiation hazard and no significant radiological threat was observed to the population of the study area.**

Keywords: fertilizer / gamma spectrometry / radium, thorium and potassium

1. Introduction

High industrialization normally occurs with intense urbanization and environmental problems of cities. Noise, air and water pollution, occupational exposure, etc., are the direct impacts of urbanization and industrialization. Many of these effects

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are not particularly harmful in isolated contacts, but continued exposure to low-level environmental pollution may be a much more serious problem for health and environmental safety (Mourad *et al.*, 2009).

Consequently, increasing attention has been paid to the many non-nuclear industries, which have the capability for low-level but consistent exposure to ionizing radiation. Such industries are capable of generating critical group or collective exposure or both; doses can be delivered both to plant workers and to populations living in the neighborhoods of plants. Cement industries, phosphate-ore industries (producing fertilizers, detergents and acids), the urea fertilizer industry, coal power plants, oil and gas extraction facilities and, to some extent, the ceramic industry are examples of these types of industries (Khan *et al.*, 1998; UNEP, 1998; Bolivar *et al.*, 2000, 2002; Righi *et al.*, 2000; Alcaraz Pelegrina and Martinez-Aguirre, 2001; Khater *et al.*, 2001; Brigden *et al.*, 2002; Saueia and Mazzilli, 2006).

As with all chemical process industries, the production of mineral fertilizers gives rise to emissions, which contribute to environmental problems, both globally and locally. Much research and expenditure has been devoted to minimizing these emissions (Santos *et al.*, 2006). Like other industries, the fertilizer industry started with plants which would be totally unacceptable today, both in terms of their waste emissions and their internal working environment. Atmospheric pollutions emitted by the fertilizer industry can include gaseous ammonia (NH_3) and ammonium salt aerosols, nitric and nitrous oxides, fluorine (as SiF_4 and HF), oxides of sulfur (SO_x), fertilizer dust, acid mists, and radiation from phosphogypsum (Butchaiah *et al.*, 2009; Hopke, 2009). Carbon dioxide is also emitted in large quantities by the fertilizer industry. Wastewater from the industry can include compounds of nitrogen, phosphate, potassium, sodium, silica, sulfur and fluorine, as well as sludge and polluted water. Solid wastes or by-products, which may or may not be reprocessed, can include phosphogypsum, calcium carbonate, soluble salts, sand and the plastic bags used to transport the fertilizer (UNEP, 1998).

Non-nuclear industries such as the fertilizer industry use raw material containing significant levels of natural radionuclides; the processing of these materials can expose the environment, workers and people living near such sites to radiation levels well above the natural background. The presence of elevated levels of Ra, Th and K in fertilizer and in environmental (soil and water) samples around a fertilizer factory complex has been established in several previous studies (Olszewska-Wasiolek, 1995; Alam *et al.*, 1997; Khan *et al.*, 1998; Righi *et al.*, 2000; Khater *et al.*, 2001; Bolivar *et al.*, 2002; Ogunleye *et al.*, 2002; El-Bahi *et al.*, 2004; Abbady *et al.*, 2005; Ahmed and El-Arabi, 2005; Saueia *et al.*, 2005; Conceicao and Bonotto, 2006; El-Arabi *et al.*, 2007; El-Aydarous, 2007; Ferdoas *et al.*, 2007;

Mourad *et al*., 2009). The general goal of many studies was to assess the environmental radioactive pollution and radiological impacts caused by a production plant of phosphate fertilizers (Alam *et al*., 1997; Ogunleye *et al*., 2002; Abbady *et al*., 2005; Ahmed and El-Arabi, 2005; Righi *et al*., 2005; Saueia and Mazzilli, 2006). Also, the assessment of fluoride and heavy metal pollution caused by such a plant was the objective of many investigations (El-Bahi *et al*., 2004; Wang *et al*., 2005; Loganathan *et al*., 2007). In particular, elevated concentrations of several natural radionuclides were found in water and soils collected from different rivers, beaches, oceans and other landscapes (UNSCEAR, 1982, 1988, 2000; Alam *et al*., 1999; Chowdhury *et al*., 1999; Ghose *et al*., 2000; Labidi *et al*., 2002; Somlai *et al*., 2002; Mireles *et al*., 2003; Cevik. *et al*., 2006; Chowdhury *et al*., 2006; Huy and Luyen, 2006; Khalifa, 2004; Mehra *et al*., 2007; Uchida *et al*., 2007; Radenkovic *et al*., 2009; Salbu and Skipperud, 2009). In the present study, to assess the impact of a urea fertilizer factory on the environment, the environmental pollution impact was analyzed by determining the concentrations of the radionuclides and other radioactive indices in soil, water and fertilizer samples which were collected from around the urea fertilizer plant, urea drain, lagoon, Shitalakhya river and the local market. Such information will lead to an accurate dosimetric evaluation of the risk of human exposure due to enhancements of TENORM (technologically-enhanced naturally occurring radioactive materials) levels.

2. Materials and Methods

2.1. Study area

The study area includes the inside and outside of a urea fertilizer factory in the district of Narsingdi which stands about 60 km away from the North-Eastern part of Dhaka on the bank of the river Sitalakhya. The basic raw material used in this factory for the production of urea is natural gas. The amount of gas used per hour is 50278 Normal m^3. 43.05% gas is used for urea processing and 56.95% used as fuel. In the first step, ammonia is produced from natural gas. Finally, urea is produced from the reaction of ammonia and carbon dioxide in certain thermodynamic conditions with the presence of a suitable catalyst. The installation capacity of this factory is 3,40,000 MT urea per year, 1137 MT urea per day and 660 MT ammonia per day.

2.2. Sampling

In order to measure the natural radioactivity in the study area, soil, water and fertilizer samples were collected from different locations of the urea fertilizer factory. Soil samples were collected from twenty-five different locations, and water

samples from fifteen different locations. The sampling locations for soil and water are inside and outside areas of the factory, the adjacent river Sitalakhya, the urea drain and the lagoon. The fertilizer samples were collected from local market. The location of sampling points are shown in Figure 1. All the soil samples were collected from an auger hole at a depth of about 15 cm from the ground. Samples were dried in an oven at a temperature of 110 $^{\circ}$C for 48 h to remove the moisture. Soil and fertilizer samples were crushed into fine powder by using a mortar and pestle. The fine quality of the samples was obtained using a 150-micron mesh size scientific sieve method. Each sample was packed and sealed in an airtight PVC container. The 15 2-l water samples were collected from the 3 locations in 2-l plastic containers. After collection, each 2-l water sample was acidified with 20 ml of 11 M HCL to prevent adsorption of radionuclides on the container walls. The collected water samples were filtered through filter paper to remove very fine undissolved particles. For radionuclide measurements, each 2-l water sample was evaporated to reduce the volume to 500 ml and packed into 150-ml cylindrical containers, sealed tightly, wrapped with thick vinyl tape around their screw necks and kept for about 4 weeks to achieve secular equilibrium between ^{222}Rn and ^{226}Ra (Tufail *et al*., 2000).

2.3. Measurement of natural radioactivity

The activity concentrations of studied samples were counted by using a 40% RE HPGe detector based on the high-resolution gamma-spectrometry system. The detector is a co-axial p-type high-purity germanium detector (Model no. GC-4020 and serial no. 07037658). The detector has a resolution of 2.0 keV and relative efficiency of 40% for 1332.5 keV gamma energy of ^{60}Co. The output of the detector is analyzed using a MCA system connected to a PC of 16384 channels. The spectral data is analyzed using the software "Genie 2000". The detector is shielded by a lead cylinder of thickness 3.7″, height 15.98″ and 15.59″ internal diameter. The upper side of the lead cylinder is covered by a lead plate of diameter 19″ and thickness 3.54″ to reduce the background level of the system. The efficiency calibration for the system is carried out using a secondary standard source in geometry available for the sample counting. Gamma transitions of 1,460 keV for ^{40}K, 295 and 352 keV of ^{214}Pb, 609 and 1120 keV of ^{214}Bi for ^{226}Ra, and 911 and 969 keV of ^{228}Ac for ^{238}Th were used for the laboratory measurement of the activity concentration of potassium, radium and thorium. The spectrum of every soil sample was collected for 50 ks.

The net count rate under the most prominent photo peaks of radium and thorium daughter peaks are calculated by subtracting the respective count rate from the background spectrum obtained for the same counting time. The

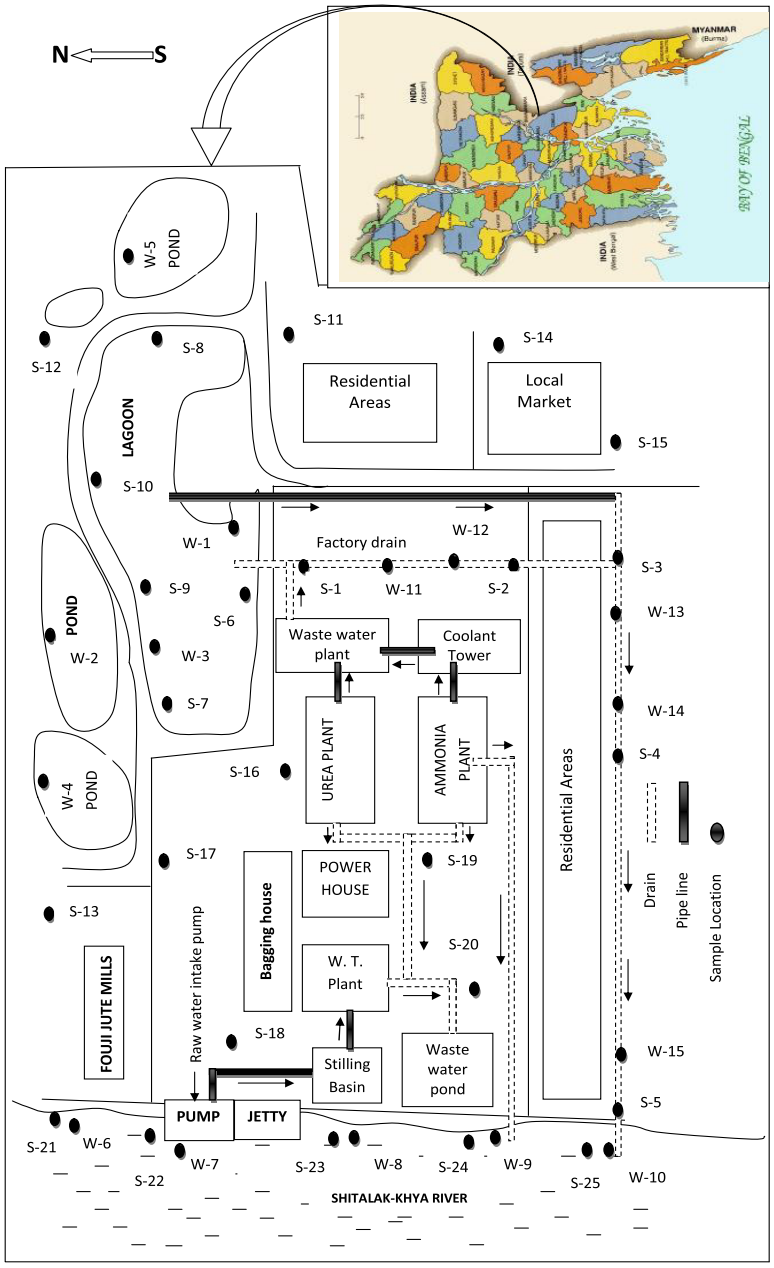

Figure 1 – ***The sampling locations of the Urea fertilizer factory.***

concentrations of radionuclides are calculated using the following equation (1) (Righi *et al.*, 2005):

$$A = \frac{cps}{E\ I\ W(\text{in kg})} \pm \sigma \quad (1)$$

where A = activity of the sample in $Bq.kg^{-1}$, cps = net counts per second (cps for sample – cps for background), E = the counting efficiency of the gamma energy = $\frac{cps}{dps\ I\ W}$, I = absolute intensity of the gamma ray and W = sample net weight.

The errors in the measurements are expressed in terms of the standard deviation (σ), which is expressed as:

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

where N_s is the sample counts measured in time T_s and N_b is the number of background counts in time T_b.

2.4. Radium equivalent activity

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in soil is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in $Bq.kg^{-1}$ to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. It is calculated through the following relation (Mehra *et al.*, 2007):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_k \quad (3)$$

where C_{Ra}, C_{Th} and C_k are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in $Bq.kg^{-1}$.

2.5. Representative level index values

To estimate the level of γ-radiation hazard associated with the natural radionuclides in the materials, the representative level index was calculated on the basis of Ra, Th and K concentration levels in the studied samples using the following equation derived by NEA-OECD (1979):

$$I\gamma r = \left(\frac{1}{150}\right)C_{Ra} + \left(\frac{1}{100}\right)C_{Th} + \left(\frac{1}{1500}\right)C_K \quad (4)$$

where C_{Ra}, C_{Th} and C_k are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in $Bq.kg^{-1}$. This index value must be less than unity in order to keep the radiation hazard insignificant, i.e., the radiation exposure due to radioactivity from soil, water and fertilizer samples is limited to unity (Mirjana *et al*., 2009).

2.6. Dose rate calculation

The total absorbed dose rate (D) in the air (outdoors) due to the uniform distribution of all the ^{226}Ra and ^{232}Th and ^{40}K in the samples 1 m above the ground surface was estimated by the formula (UNSCEAR, 1988):

$$D = 0.427C_{Ra} + 0.662C_{Th} + 0.0432C_k \ (nGy.h^{-1}) \quad (5)$$

where C_{Ra}, C_{Th} and C_k are, respectively, the average activity concentrations of Ra, Th and K contents in samples in $Bq.kg^{-1}$.

3. Results and discussion

3.1. Soil and water samples

The activity concentrations determined for the radionuclides in the analyzed soil samples are listed in Table I. The presented results of the contents of radionuclides in soil samples, taken from the urea drain and inside the factory, show low activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K. The minimum activity concentration, 3.16 ± 0.32 $Bq.kg^{-1}$ of ^{226}Ra, was determined in the soil sample from inside the factory, while the maximum value of 10.28 ± 0.55 $Bq.kg^{-1}$ was found in the soil from Sitalakhya river. The activity concentrations of ^{232}Th were in the range 4.89 ± 0.45–15.82 ± 0.45 $Bq.kg^{-1}$, with a minimum value for inside the factory and a maximum for the Sitalakhya river. The content of ^{40}K had the lowest value of 24.96 ± 0.23 $Bq.kg^{-1}$ for the soil sample from inside the factory and relatively higher values up to 60.49 ± 0.56 $Bq.kg^{-1}$ for the soils from the urea drain, and inside and outside the factory.

These results are comparatively lower in accordance with previous studies in Egypt (21.5 (15.4–33.8), 14.2 (10.4–19.3), 219 (128–281) $Bq.kg^{-1}$) (Mourad *et al*., 2009), the Bangladeshi river Karnaphuli 35.9 ± 18.9 (18.4 ± 2.6–85.2 ± 8.4), 65.5 ± 12.2 (50.8 ± 4.2–88.4 ± 7.7), 272 ± 35 (217 ± 15.5–320 ± 21) $Bq.kg^{-1}$) and Sangu (27.8 ± 2.9 (31.9 ± 3.7–24 ± 3.0), 57.5 ± 3.0 (52.4 ± 5.5–61.7 ± 6.7), 255 ± 18 (212 ± 15–292 ± 18) $Bq.kg^{-1}$) and the coastal soil of Chittagong (34.6 ± 20.8 (14.0 ± 0.5–89.2 ± 6.4), 60 ± 29.2 (28.3 ± 2.4–129 ± 10.4), 438 ± 142 (200 ± 22–772 ± 33) $Bq.kg^{-1}$) (Chowdhury *et al*., 1999), a southern district of Bangladesh (42, 81, 833 $Bq.kg^{-1}$) (Chowdhury *et al*., 2006), the Um Sleimat area of Egypt

TABLE I
Measurement of radioactivity in soil samples.

Sample locations with sample ID	Radium concentration in Soil, C_{Ra}(Bq.kg^{-1})	Thorium concentration in Soil, C_{Th} (Bq.kg^{-1})	Potassium concentration in Soil, C_K (Bq.kg^{-1})	Activity Ratio		Radium equivalent activity, Ra_{eq}(Bq.kg^{-1})	Representative Level Index ($I_{\gamma r}$) (Bq.kg^{-1})	Absorbed Dose Rate (nGy.h^{-1})
				^{226}Ra/^{232}Th	^{226}Ra/^{40}K			
Urea drain								
Soil-1	4.28 ± 0.36	5.96 ± 0.24	57.47 ± 0.75	0.72	0.07	17.23 ± 0.76	0.126 ± 0.0053	8.26 ± 0.35
Soil-2	4.62 ± 0.48	6.23 ± 0.43	56.84 ± 0.66	0.74	0.08	17.90 ± 1.15	0.131 ± 0.0079	8.55 ± 0.52
Soil-3	5.08 ± 0.22	5. 58 ± 0.32	57.12 ± 0.59	0.91	0.09	17.46 ± 0.72	0.128 ± 0.0051	8.33 ± 0.33
Soil-4	4.04 ± 0.52	5.95 ± 0.38	60.02 ± 0.81	0.68	0.07	17.17 ± 1.13	0.126 ± 0.0078	8.26 ± 0.51
Soil-5	5.02 ± 0.29	6.03 ± 0.18	59.71 ± 0.73	0.83	0.08	18.24 ± 0.60	0.134 ± 0.0042	8.71 ± 0.27
Lagoon								
Soil-6	7.68 ± 0.47	9.48 ± 0.44	54.11 ± 0.71	0.81	0.14	25.40 ± 1.15	0.182 ± 0.0080	11.9 ± 0.52
Soil-7	8.03 ± 0.41	10.38 ± 0.67	51.67 ± 0.69	0.77	0.15	26.85 ± 1.42	0.192 ± 0.0099	12.5 ± 0.65
Soil-8	7.49 ± 0.35	7.76 ± 0.54	60.03 ± 0.49	0.97	0.12	23.21 ± 1.16	0.168 ± 0.0081	10.9 ± 0.53
Soil-9	10.18 ± 0.24	11.98 ± 0.78	54.98 ± 0.75	0.85	0.19	31.54 ± 1.41	0.224 ± 0.0099	14.7 ± 0.65
Soil-10	5.72 ± 0.38	10.96 ± 0.65	48.43 ± 0.66	0.52	0.12	25.12 ± 1.36	0.180 ± 0.0095	11.8 ± 0.62
Outside of the factory								
Soil-11	7.72 ± 0.55	10.35 ± 0.43	54.93 ± 0.73	0.75	0.14	26.75 ± 1.22	0.192 ± 0.0085	12.5 ± 0.55
Soil-12	9.92 ± 0.39	9.89 ± 0.52	51.24 ± 0.61	1.00	0.19	28.00 ± 1.18	0.199 ± 0.0082	13.0 ± 0.54
Soil-13	4.98 ± 0.45	12.18 ± 0.64	55.62 ± 0.27	0.41	0.09	26.68 ± 1.39	0.192 ± 0.0096	12.6 ± 0.63
Soil-14	7.43 ± 0.63	10.59 ± 0.33	53.46 ± 0.69	0.70	0.14	26.69 ± 1.16	0.191 ± 0.0080	12.5 ± 0.52
Soil-15	6.57 ± 0.58	7.79 ± 0.23	60.49 ± 0.56	0.84	0.11	22.37 ± 0.95	0.162 ± 0.0065	10.6 ± 0.42

TABLE I
Continued.

Sample locations with sample ID		Radium concentration in Soil, C_{Ra} (Bq.kg^{-1})	Thorium concentration in Soil, C_{Th} (Bq.kg^{-1})	Potassium concentration in Soil, C_K (Bq.kg^{-1})	Activity Ratio		Radium equivalent activity, Ra_{eq} (Bq.kg^{-1})	Representative Level Index ($I_{\gamma r}$) (Bq.kg^{-1})	Absorbed Dose Rate (nGy.h^{-1})
					$^{226}Ra/^{232}Th$	$^{226}Ra/^{40}K$			
Inside of the factory									
Soil-16		4.76 ± 0.38	6.02 ± 0.28	27.29 ± 0.37	0.71	0.17	15.47 ± 0.89	0.110 ± 0.0056	7.20 ± 0.36
Soil-17		5.58 ± 0.41	5.97 ± 0.33	24.96 ± 0.23	0.93	0.22	16.04 ± 0.90	0.114 ± 0.0062	7.41 ± 0.40
Soil-18		7.48 ± 0.29	4.89 ± 0.45	26.89 ± 0.35	1.53	0.28	16.54 ± 0.96	0.117 ± 0.0067	7.59 ± 0.44
Soil-19		4.51 ± 0.56	7.85 ± 0.26	37.53 ± 0.48	0.57	0.12	18.63 ± 0.97	0.134 ± 0.0067	8.74 ± 0.43
Soil-20		3.16 ± 0.32	6.36 ± 0.19	27.38 ± 0.34	0.49	0.16	14.36 ± 0.62	0.103 ± 0.0043	6.74 ± 0.28
Sitalakhya River									
Soil-21		5.09 ± 0.48	11.67 ± 0.50	39.25 ± 0.53	0.44	0.13	24.80 ± 1.24	0.177 ± 0.0085	11.6 ± 0.56
Soil-22		10.28 ± 0.55	10.23 ± 0.21	36.87 ± 0.59	1.00	0.28	27.75 ± 0.90	0.195 ± 0.0062	12.8 ± 0.40
Soil-23		4.96 ± 0.39	11.55 ± 0.56	32.25 ± 0.52	0.43	0.15	23.96 ± 1.23	0.170 ± 0.0085	11.2 ± 0.56
Soil-24		3.63 ± 0.51	15.82 ± 0.45	40.46 ± 0.43	0.23	0.09	29.37 ± 1.19	0.209 ± 0.0082	13.8 ± 0.53
Soil-25		5.12 ± 0.43	9.63 ± 0.61	34.11 ± 0.23	0.53	0.15	21.52 ± 1.32	0.153 ± 0.0091	10.0 ± 0.60
Mean		6.13 ± 0.43	8.84 ± 0.42	46.52 ± 0.56	0.74	0.14	22.36 ± 1.08	0.160 ± 0.0075	10.5 ± 0.49
Median		5.12 ± 0.43	9.48 ± 0.44	51.67 ± 0.69	0.75	0.13	23.21 ± 1.16	0.168 ± 0.0081	10.9 ± 0.53
Range	**Max**	10.28 ± 0.55	15.82 ± 0.45	60.49 ± 0.56	1.53	0.28	29.37 ± 1.19	0.209 ± 0.0082	13.8 ± 0.53
	Min	3.16 ± 0.32	4.89 ± 0.45	24.96 ± 0.23	0.23	0.07	14.36 ± 0.62	0.103 ± 0.0043	6.74 ± 0.28

(77.9 ± 8.7 (45.5 ± 7–130 ± 11), 52.8 ± 7.2 (36.7 ± 6–77.5 ± 9), 1424.9 ± 38 (1299.7 ± 36–1521.6 ± 39) Bq.kg^{-1}) (El-Arabi *et al.*, 2007)), Taif in Saudi Arabia (23.8 ± 2.4 (13 ± 1.2–33 ± 3.4), 18.6 ± 1.7 (11 ± 1–27 ± 4.2), 162.8 ± 7.6 (129 ± 5.7–230 ± 11) Bq.kg^{-1}) (El-Aydarous, 2007), the South-East of Vietnam (19.6, 31, 34.6 Bq.kg^{-1}) (Huy and Luyen, 2006), and Mexico (Zacatecas and Guandalupe) (23, 19, 530 Bq.kg^{-1}) (Mireles *et al.,* 2003), as well as world quoted values for soils (25 (7–50), 25 (10–50) and 370 (100–700) Bq.kg^{-1}) (Mirjana *et al.*, 2009) for ^{226}Ra, ^{232}Th and ^{40}K, respectively. Therefore, it can be concluded that due to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the soil, none of the studied samples is considered a radiological hazard and this area is safe.

A comparison of radionuclide concentrations in the water of the three regions in the study area is shown in Table II. The water sample collected from the urea drain shows a higher content of ^{226}Ra (7.36 ± 0.42 Bq.L^{-1}) than the other two regions. The maximum amounts of ^{232}Th (8.59 ± 0.37 Bq.L^{-1}) and ^{40}K (35.48 ± 0.24 Bq.L^{-1}) were found in the water samples collected from the lagoon beside the urea fertilizer factory. The lowest concentrations of ^{226}Ra (1.22 ± 0.41 Bq.L^{-1}), ^{232}Th (1.21 ± 0.06 Bq.L^{-1}) and ^{40}K (7.48 ± 0.53 Bq.L^{-1}) were found in the water samples collected from Sitalakhya river.

A comparative study of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in water samples of the study area with the previous measurements from different countries of the world shows the average activities of ^{226}Ra are higher than those observed in Egypt (0.58 Bq.L^{-1}) (Mourad *et al.*, 2009), the Chittagong Region of Bangladesh (drinking water: 0.043 ± 0.017 Bq.L^{-1}) (Alam *et al.*, 1999), the Eastern Black Sea region of Turkey (tap water: 0.019 Bq.L^{-1}) (Cevik *et al.*, 2006), Qena, Egypt (drinking water: 0.048 ± 0.0259 Bq.L^{-1}, groundwater: 0.07918 ± 0.02886 Bq.L^{-1}), Safaga-Quseir, Egypt (drinking water: 0.05624 ± 0.02849 Bq.L^{-1}, groundwater: 0.11285 ± 0.0333 Bq.L^{-1}) (Khalifa, 2004), Tunisia (springs: 0.03404 – 3.8961 Bq.L^{-1}) (Labidi *et al.*, 2002) and Hungary (bottled mineral water: 0.0999 to 2.997 Bq.L^{-1}) (Somlai *et al.*, 2002), while they are lower than those found in the Portuguese rivers Tejo and Duro (16.2 ± 6.7 and 19.8 ± 8.7 Bq.L^{-1}), and the Bay of Bengal (surface seawater: 5.4 ± 2.4 to 29.0 ± 8.3 Bq.L^{-1}) (Ghose *et al.*, 2000). A very high concentration of ^{226}Ra was reported in the Ikaria Sea (1200 ± 1000 Bq.L^{-1}), French rivers (1 to 730 Bq.L^{-1}) and Czechoslovakia (10 to 3800 Bq.L^{-1}) (Ghose *et al.*, 2000). On the other hand, the average activity of this radionuclide in the area also closely agrees with the Chinese river, Yangtze (2.8–8.9 Bq.L^{-1}), a Greek region (1.5 ± 0.3–1.7 ± 0.4 Bq.L^{-1}) and the UK (1.3 – 3.1 Bq.L^{-1}) (Ghose *et al.*, 2000). The measured activity concentrations of ^{232}Th (3.82 ± 0.24 Bq.L^{-1}) in the same water samples are higher than the previous results from Egypt (0.05 Bq.L^{-1}) (Mourad *et al.*, 2009), the Chittagong Region of Bangladesh (drinking water: 0.19 ± 0.05 Bq.L^{-1}) (Alam *et al.*, 1999), Qena, Egypt

TABLE II
Measurement of radioactivity in water samples.

Sample locations with sample ID		Radium concentration in water, C_{Ra} (Bq.L^{-1})	Thorium concentration in water, C_{Th} (Bq.L^{-1})	Potassium concentration in water, C_K (Bq.L^{-1})	Activity Ratio		Radium equivalent activity, Ra_{eq} (Bq.L^{-1})	Representative Level Index ($I\gamma r$) in Bq.L^{-1}	Absorbed Dose Rate (nGy.h^{-1})
					$^{226}Ra/^{232}Th$	$^{226}Ra/^{40}K$			
Lagoon									
Water-1		3.09 ± 0.24	2.67 ± 0.16	26.24 ± 0.35	1.15	0.12	8.929 ± 0.50	0.065 ± 0.0034	2.9 ± 0.22
Water-2		6.45 ± 0.29	5.21 ± 0.08	35.48 ± 0.24	1.24	0.18	16.63 ± 0.42	0.119 ± 0.0029	7.74 ± 0.19
Water-3		2.86 ± 0.45	8.59 ± 0.37	20.57 ± 0.40	0.33	0.14	16.73 ± 1.01	0.119 ± 0.0070	7.8 ± 0.45
Water-4		3.42 ± 0.52	4.51 ± 0.29	19.16 ± 0.17	0.76	0.18	11.34 ± 0.95	0.081 ± 0.0065	5.27 ± 0.42
Water-5		4.16 ± 0.36	1.63 ± 0.43	24.39 ± 0.15	2.55	0.17	8.369 ± 0.99	0.060 ± 0.0068	3.91 ± 0.44
Sitalakhya river									
Water-6		1.22 ± 0.41	1.21 ± 0.06	9.59 ± 0.13	1.00	0.13	3.689 ± 0.51	0.026 ± 0.0034	1.74 ± 0.22
Water-7		3.56 ± 0.32	3.29 ± 0.16	12.65 ± 0.32	1.08	0.28	9.239 ± 0.57	0.065 ± 0.0039	4.24 ± 0.26
Water-8		1.27 ± 0.25	2.98 ± 0.21	7.48 ± 0.53	0.43	0.17	6.107 ± 0.59	0.043 ± 0.0041	2.84 ± 0.27
Water-9		2.65 ± 0.45	4.16 ± 0.09	20.53 ± 0.24	0.64	0.13	10.18 ± 0.60	0.073 ± 0.0041	4.77 ± 0.26
Water-10		3.02 ± 0.14	1.23 ± 0.11	19.38 ± 0.17	2.46	0.16	6.271 ± 0.31	0.045 ± 0.0021	2.94 ± 0.14
Urea drain									
Water-11		2.82 ± 0.37	2.11 ± 0.10	13.00 ± 0.17	1.33	0.21	6.838 ± 0.53	0.049 ± 0.0036	3.16 ± 0.23
Water-12		4.95 ± 0.29	1.98 ± 0.49	24.29 ± 0.39	2.50	0.20	9.652 ± 1.02	0.069 ± 0.0071	4.47 ± 0.47
Water-13		5.57 ± 0.33	6.14 ± 0.26	17.32 ± 0.56	0.91	0.32	15.68 ± 0.74	0.110 ± 0.0052	7.19 ± 0.34
Water-14		6.85 ± 0.51	8.36 ± 0.41	27.86 ± 0.43	0.82	0.25	20.95 ± 1.13	0.148 ± 0.0078	9.66 ± 0.51
Water-15		7.36 ± 0.42	3.19 ± 0.31	11.59 ± 0.32	2.31	0.64	12.81 ± 0.89	0.089 ± 0.0061	5.76 ± 0.40
Mean		3.95 ± 0.36	3.82 ± 0.24	19.30 ± 0.30	1.30	0.22	10.90 ± 0.72	0.077 ± 0.0049	5.05 ± 0.32
Median		3.42 ± 0.52	3.19 ± 0.31	19.38 ± 0.17	1.08	0.18	9.652 ± 1.02	0.069 ± 0.0071	4.47 ± 0.47
Range	**Max**	7.36 ± 0.42	8.59 ± 0.37	35.48 ± 0.24	2.55	0.64	20.95 ± 1.13	0.148 ± 0.0078	9.66 ± 0.51
	Min	1.22 ± 0.41	1.21 ± 0.06	7.48 ± 0.53	0.33	0.12	3.689 ± 0.51	0.026 ± 0.0034	1.74 ± 0.22

(drinking water: 0.02738 ± 0.01628 $Bq.L^{-1}$, groundwater: 0.0407 ± 0.02294 $Bq.L^{-1}$) and Safaga-Quseir, Egypt (drinking water: 0.02923 ± 0.01665 $Bq.L^{-1}$, groundwater: 0.05143 ± 0.0222 $Bq.L^{-1}$) (Khalifa, 2004). The concentration observed for ^{40}K is higher than that of the reported value from Egypt (0.05 $Bq.L^{-1}$) (Mourad *et al.*, 2009), the Eastern Black Sea region of Turkey (tap water: 0.019 $Bq.L^{-1}$) (Cevik *et al.*, 2006) and the Chittagong Region of Bangladesh (drinking water: 4.16 ± 1.58 $Bq.L^{-1}$) (Alam *et al.*, 1999).

The gamma-ray radiation hazards due to the specified radionuclides were also assessed by the indices, i.e., the radium equivalent activity (Ra_{eq}), the activity ratio, the representative level index (Iγr) and the absorbed dose rate. The results obtained for the soils and waters studied in this work are presented in Tables I and II. The presented results show that the Ra_{eq} index for the soil samples had values in the range 14.36 ± 0.62 $Bq.kg^{-1}$ (inside the fertilizer factory) to 29.37 ± 1.19 $Bq.kg^{-1}$ (Sitalakhya river) compared with the population-weighted average value of global primordial radiation of 59 $Bq.kg^{-1}$ (Mirjana *et al.*, 2009). For water samples the Ra_{eq} ranges from 3.689 ± 0.51 to 20.95 ± 1.13 $Bq.L^{-1}$. The index value Iγr must be less than unity in order to keep the radiation hazard insignificant (Ferdoas *et al.*, 2007). The calculated values of Iγr were within the limit value of 0.209 ± 0.0082–0.103 ± 0.0043 $Bq.kg^{-1}$ for soil and 0.026 ± 0.0034 to 0.148 ± 0.0078 $Bq.L^{-1}$ for water. The values of absorbed dose rates in samples range from 6.74 ± 0.28–13.8 ± 0.53 $nGy.h^{-1}$ with a mean of 10.5 ± 0.49 $nGy.h^{-1}$ for soil and 9.66 ± 0.51–1.74 ± 0.22 $nGy.h^{-1}$ with a mean of 5.05 ± 0.32 $nGy.h^{-1}$ for water, *i.e.*, the measured values are less than the world average of 55 $nGy.h^{-1}$ (UNSCEAR, 1998). The $^{226}Ra/^{232}Th$ activity ratio varies in the ranges 0.33–2.55 for water samples, and 0.23–1.53 for soil. The $^{226}Ra/^{40}K$ activity ratios are in the ranges 0.07–0.28 for soils, and 0.12–0.64 for water. Therefore, it can be concluded that the surface radiation dose for the study area is within the global range.

3.2. Fertilizer samples

The measured radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and the values of radiation hazard parameters in different fertilizers are shown in Table III. The radioactivity of ^{226}Ra in different fertilizers ranged from 3.55 ± 0.33 to 90.65 ± 3.17 $Bq.kg^{-1}$. Sulfur (S), MOP (muriate of potash) (KCl), TSP (triple superphosphate) [$Ca(H_2PO_4)_2$. H_2O] and DAP (diammonium phosphate) [$(NH_4)_2PO_4$] contained high contents of ^{226}Ra. Urea is produced from natural gas, nitrogen and water; the presence of small amounts of ^{226}Ra, ^{232}Th and ^{40}K may be due to impurities present in the raw materials. The radioactivity of ^{226}Ra in TSP was 90.65 ± 3.17 $Bq.kg^{-1}$, in DAP 46.46 ± 2.32 $Bq.kg^{-1}$, in MOP 4.90 ± 0.39 $Bq.kg^{-1}$, and in sulfur 55.20 ± 2.15 $Bq.kg^{-1}$. The ^{226}Ra, ^{232}Th and ^{40}K concentrations determined

TABLE III
Measurement of radioactivity in fertilizer samples collected from the local market.

Fertilizer samples collected from local market		Radium Concentration, C_{Ra} (Bq.kg^{-1})	Thorium concentration, C_{Th} (Bq.kg^{-1})	Potassium concentration, C_K(Bq.kg^{-1})	Activity Ratio		Radium equivalent activity, Ra_{eq} (Bq.kg^{-1})	Representative Level Index (Iγr) in Bq.kg^{-1}	Absorbed Dose Rate (nGy.h^{-1})
					$^{226}Ra/^{232}Th$	$^{226}Ra/^{40}K$			
Urea		3.55 ± 0.33	4.76 ± 0.25	3.55 ± 0.05	0.74	1.00	10.63 ± 0.69	0.074 ± 0.0047	4.82 ± 0.31
Sulphur		55.20 ± 2.15	17.20 ± 0.82	51.46 ± 0.71	3.21	1.07	83.76 ± 3.38	0.574 ± 0.0230	37.18 ± 1.49
MOP (muriate of potash)		4.90 ± 0.39	9.35 ± 0.57	3051.71 ± 19.53	0.52	0.002	253.25 ± 2.71	2.162 ± 0.0213	140.12 ± 1.39
TSP (triple superphosphate)		90.65 ± 3.17	26.38 ± 1.40	83.46 ± 1.12	3.44	1.09	134.80 ± 5.26	0.924 ± 0.0358	59.78 ± 2.33
DAP (diammonium phosphate)		46.46 ± 2.32	17.94 ± 0.93	76.78 ± 1.01	2.59	0.61	78.03 ± 3.73	0.540 ± 0.0254	35.03 ± 1.65
Mean		40.15 ± 1.67	15.13 ± 0.794	653.39 ± 4.484	2.10	0.75	112.10 ± 3.15	0.855 ± 0.0221	55.38 ± 1.43
Median		46.46 ± 2.32	17.20 ± 0.82	76.78 ± 1.01	2.59	1	83.76 ± 3.38	0.574 ± 0.0230	37.18 ± 1.49
Range	Max	90.65 ± 3.17	26.38 ± 1.40	3051.71 ± 19.53	3.44	1.09	253.25 ± 2.71	2.162 ± 0.0213	140.12 ± 1.39
	Min	3.55 ± 0.33	4.76 ± 0.25	3.55 ± 0.05	0.52	0.002	10.63 ± 0.69	0.074 ± 0.0047	4.82 ± 0.31

in this work for urea fertilizer (3.55 ± 0.33, 4.76 ± 0.25 and 3.55 ± 0.05 Bq.kg^{-1}, respectively) were found to be lower than those reported for TSP (90.65 ± 3.17, 26.38 ± 1.40 and 83.46 ± 1.12 Bq.kg^{-1}), DAP (46.46 ± 2.32, 17.94 ± 0.93 and 76.78 ± 1.01 Bq.kg^{-1}), MOP (4.90 ± 0.39, 9.35 ± 0.57 and 3051.71 ± 19.53 Bq.kg^{-1}) and sulfur (55.20 ± 2.15, 17.20 ± 0.82, 51.46 ± 0.71 Bq.kg^{-1}) for the same radionuclides, which agreed with Alam *et al.* (1997) (5.4 ± 1.5, 3.4 ± 1.7 and 7.9 ± 2.4 Bq.kg^{-1} in urea). Hence, the probability of increasing the radioactivity level in soil is meager when using the urea fertilizer in cultivation rather than TSP, DAP, MOP, sulfur, phosphogypsum and single superphosphate.

A review of the worldwide data on natural radioactivity in fertilizers was presented in UNSCEAR reports (1988, 2000). The concentrations of ^{226}Ra, ^{232}Th and ^{40}K of the present study (90.65 ± 3.17, 26.38 ± 1.40 and 83.46 ± 1.12 Bq.kg^{-1}) are comparable with the reported concentrations of the same radionuclides in TSP fertilizer in Egypt (Mourad *et al.*, 2009) (516 ± 23, 6.5 ± 3.6 and 107 ± 20 Bq.kg^{-1}), Pakistan (Khan *et al.*, 1998) (558.6, 84.8 and 143 Bq.kg^{-1}), Brazil (Saueia *et al.*, 2005) (122 ± 6, 538 ± 36 and 147 ± 17 Bq.kg^{-1}) and Bangladesh (Alam *et al.*, 1997) (323.8 ± 24.4, 22.0 ± 2.8 and 241.0 ± 4.2 Bq.kg^{-1}).

The average activity of ^{232}Th in the different fertilizers ranged from 4.76 ± 0.25 to 26.38 ± 1.40 Bq.kg^{-1}; the highest activity was observed in TSP fertilizer and the lowest was observed in urea fertilizer. The ^{232}Th activity of the present study in phosphate fertilizers is very much lower than the value reported by Conceicao and Bonotto (2006) and Saueia *et al.* (2005) and exceeds the values reported in Egypt (Mourad *et al.*, 2009), Israel (Sam and Holm, 1995), and Algeria, Jordan, Tunisia and Sudan (Olszewska-Wasiolek, 1995). The activity of ^{40}K was found to be high in the fertilizer muriate of potash (MOP), which contained a ^{40}K activity of 3051.71 ± 19 Bq.kg^{-1}. The activity of ^{40}K in TSP fertilizer (83.46 ± 1.12Bq.kg^{-1}) is lower than the values in Egypt (Mourad *et al.*, 2009), Bangladesh (Alam *et al.*, 1997) and Pakistan (Khan *et al.*, 1998).

The isotope ratios of the fertilizer samples (mean value of Tab. III) and the soil samples (mean value of Tab. I) for the measured radionuclides ^{226}Ra, ^{232}Th and ^{40}K are 0.15, 0.58 and 0.08. The data confirms that the fertilizer plant has very low radiological consequences on the surroundings.

The absorbed dose rates were in the range 4.82–140.12 nGy.h^{-1} with an average value of 55.38 nGy.h^{-1} for fertilizer samples. The value given for the absorbed dose rate is 44 nGy.h^{-1}, while a higher value of 55 nGy.h^{-1} (UNSCEAR, 1982), (24–85 nGy.h^{-1}) is the cited value for terrestrial radiation in 23 countries worldwide (UNSCEAR, 1988). The UNSCEAR report gave a wider range (18–93 nGy.h^{-1}) with an average value of 59 nGy.h^{-1} (UNSCEAR, 2000). The

results showed that the absorbed dose rates for the fertilizer samples are comparable with the world limits. The Ra_{eq} concentration in fertilizer samples determined in this study showed a wide range (10.63–253.25 $Bq.kg^{-1}$) and other reported values by Ogunleye *et al*. (2002) (584 and 1095.9 $Bq.kg^{-1}$), Ahmed and El-Arabi (2005) (461.7 $Bq.kg^{-1}$) exceed this range. The Ra_{eq} concentration determined in this work for fertilizer is lower than that reported by El-Bahi *et al*. (2004) (992.36 and 1005.84 $Bq.kg^{-1}$) and Saueia *et al*. (2005) (902.66 $Bq.kg^{-1}$). The representative level index ranged from 0.074 to 2.162 $Bq.kg^{-1}$ with an average value of 0.855 $Bq.kg^{-1}$ for fertilizer samples, which is lower than that of Khan *et al*. (1998) (12.12 $Bq.kg^{-1}$) and Saueia *et al*. (2005) (7.92 $Bq.kg^{-1}$). The activity ratio of $^{226}Ra/^{232}Th$ ranged from 0.52 to 3.44 with a mean of 2.10, and the ratio for $^{226}Ra/^{40}K$ ranged from 0.002 to 1.09 with a mean of 0.75 in different fertilizers. These values are within the range of Mourad *et al*. (2009) and Khan *et al*. (1998) and exceed the values reported in Saueia *et al*. (2005).

4. Conclusion

The results showed that the ^{226}Ra, ^{232}Th and ^{40}K concentrations in all types of studied environmental and fertilizer samples are lower compared with the maximum permissible limit set by various international organizations and the results of other studies, with a few exceptions. We therefore conclude that the study area does not pose any radiological hazard. The data presented in this study will also serve as a baseline survey of primordial radionuclide concentrations in the study area.

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