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Research Article

Comparison of Natural Radioactivity of Commonly Used Fertilizer Materials in Egypt and Japan

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Specific activities of ^{238}U, ^{232}Th, and ^{40}K in the environment have been redistributed by the use of fertilizers in agriculture so their concentrations in fertilizer materials should be measured to identify the safe utilization of fertilizers. In the present work, the specific activities of these radionuclides in five commonly used fertilizers in Egypt and five fertilizers used in Japan were measured by HPGe and γ-ray spectrometry. The average values of ^{238}U, ^{232}Th, and ^{40}K in Japanese fertilizers were less than their values in Egyptian fertilizers but both had some samples with specific activities greater than the recommended limiting values. The radiological hazards of radium equivalent activity (Ra_{eq}), external (H_{ex}) and internal (H_{in}) indexes, alpha and gamma indexes, and annual effective dose, due to the presence of these radionuclides, were calculated and compared with each other.

1. Introduction

Fertilizers play an important role in the agriculture sector to increase crop yields so fertilizer industries have spread out all over the world. Fertilizers are composed mainly of nitrogen (N), phosphorus (P), and potassium (K), which are essential elements for plants growth. The phosphorus portion is taken from phosphate rocks, which contain a relatively high concentration of naturally occurring ^{238}U, ^{232}Th, and ^{40}K and their radioactive daughters [1, 2]. Therefore, natural radioactivity in soil varies from one location to another due to the extensive use of fertilizer which is the main source of radioactivity in soil other than its natural origin [3, 4]. The extensive use of fertilizers can increase the amount of radionuclides in soils and in groundwater and consequential ingestion by humans through exposure routes such as drinking water and the food chain [4, 5]. Once deposited in bone tissue ^{226}Ra has a high potential for causing biological damage because of the continuous irradiation of the human skeleton over many years and it can induce bone sarcoma [4, 5].

Factory workers that produce fertilizers and those who use fertilizers in agriculture are exposed to gamma radiation (external exposure) and alpha particles (internal exposure) emitted from the radionuclides of the ^{238}U series, ^{232}Th series, and ^{40}K. External exposure occurs directly by γ-rays, whereas internal exposure occurs by α-particles that result from the inhalation of radon and its progenies. Consequently, the α-particle dose is delivered directly to the bronchial tissue, creating a potential for radiogenic lung cancer [6–8]. Therefore, radiation released from fertilizers has a potential of causing cancers in individuals exposed to significant levels so that monitoring of natural radioactivity in fertilizers has an importance from the viewpoint of radiation protection [9, 10].

There is a significantly increasing international awareness of radiation hazards of fertilizer materials as a potential source of risk to workers, members of the public, and the environment. Japan as a developed country has a good radiation protection system so that, in the present work, the natural specific activities of ^{238}U, ^{232}Th, and ^{40}K, in commonly used

Table 1: Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Egyptian fertilizers.

Samples name	Sample code	Composition of fertilizer	Specific activity (Bq kg^{-1})		
			^{226}Ra	^{232}Th	^{40}K
Fertilizer 1	EF-1	SSP {P_2O_5 (15%)}	761 ± 31	67 ± 13	251 ± 94
Fertilizer 2	EF-2	SSP {P_2O_5 (12%)}	557 ± 19	15 ± 6	175 ± 27
Fertilizer 3	EF-3	SSP {P_2O_5 (16%)}	782 ± 24	14 ± 8	222 ± 24
Fertilizer 4	EF-4	NPK fertilizer	443 ± 11	ND	88 ± 22
Fertilizer 5	EF-5	Triple fertilizer	312 ± 14	ND	175 ± 31

ND: nondetectable value (the measured value less than thorium detection limit of 3 Bq kg^{-1}).

Table 2: Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Japanese fertilizers.

Samples name	Sample code	Composition of fertilizer	Specific activity (Bq kg^{-1})		
			^{226}Ra	^{232}Th	^{40}K
Fertilizer 1	JF1	Phosphoric acid 10%	25 ± 1	5 ± 2	3909 ± 21
Fertilizer 2	JF2	Phosphoric acid 8%	62 ± 1	15 ± 1	3280 ± 17
Fertilizer 3	JF3	Phosphoric acid 20%	74 ± 1	6 ± 1	48 ± 2
Fertilizer 4	JF4	Phosphoric acid 20%	200 ± 2	12 ± 1	231 ± 4
Fertilizer 5	JF5	Phosphoric acid 17%	1264 ± 5	8 ± 2	31 ± 5

fertilizer materials in Egypt, were measured and their values were compared to those of Japanese fertilizers. In addition, the radiological hazards of radium equivalent activity (Ra_{eq}), external (H_{ex}) and internal (H_{in}) indexes, alpha and gamma indexes, and annual effective dose, due to the presence of those radionuclides, were calculated and compared with recommended limits.

2. Materials and Methods

2.1. Sample Preparation. Samples of five commonly used fertilizers in Egypt and five fertilizers used in Japan as well were collected from different companies and factories, as shown in Tables 1 and 2. The selected samples were crushed and sieved through a 1 mm mesh size to remove the larger grains size and to become more uniform. Then, these samples were oven dried in a temperature controlled oven at 110°C for 24 h to ensure that the moisture is completely removed. After moisture removal, the samples were cooled down to room temperature in a desiccator.

The dried homogenized samples were packed into airtight polyethylene containers (6 cm in diameter and 8 cm in height). The containers were carefully sealed with adhesive epoxy to prevent ^{222}Rn and ^{220}Rn from escaping. Each sample was stored in its sealed container for four weeks to achieve radioactive secular equilibrium. A similarly sealed empty container of the same geometry was left for the same time period in order to measure the radionuclide background [15].

2.2. Measurement of Radionuclide Activities with γ-Ray Spectrometry. Specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in the samples were measured using an HPGe detector, ORTEC (model: GMX-70230 EG&G) with a volume of 190 cm^3, a measured efficiency of 70%, and an energy resolution of 2.3 keV at 1332.5 keV. This was connected to a personal computer with a data acquisition system that has a Multichannel analyzer, model CANBERRA Multi Port II (4,096 channels). The data analysis was carried out via an APTEC MCA software program.

The HPGe detector's peak efficiency was determined using standard point sources of ^{60}Co, ^{133}Ba, ^{137}Cs, ^{22}Na, and a standard source of ^{226}Ra, maintained in the same container geometry as that used for the samples. Since radium (^{226}Ra) and its progenies produce 98.5% of the radiological effects of the uranium series, the activities of ^{238}U and the precursors of ^{226}Ra are normally ignored. Therefore, the reference for the ^{238}U series is often ^{226}Ra instead of ^{238}U [15]. The ^{226}Ra activity was deduced from the γ-rays of energies of 351.9 keV associated with the decay of ^{214}Pb and 609.3 keV γ-rays associated with the decay of ^{214}Bi. The 186 keV photon peak of ^{226}Ra was not used because of the interfering peak of ^{235}U with energy of 185.7 keV. The ^{232}Th concentration was estimated from the γ-rays of energies of 911.1 keV associated with the decay of ^{228}Ac and the 583.4 keV line associated with the decay of ^{208}Tl. The ^{40}K concentration was obtained from 1460.8 keV γ-rays from the decay of ^{40}K itself [16–21]. The specific activity concentration (Bq kg^{-1}) of those radionuclides was calculated from (1) [15]. For the calculations of specific radioactivity, coincidence-summing and self-absorption correction factors have not been applied.

$$A = \frac{C}{p w t \varepsilon}, \tag{1}$$

where C is the net counts above the background, p is the absolute emission probability of the γ-ray decay, w is the net dry sample weight (kg), t is the measurement time (s), and ε is the absolute efficiency of the detector.

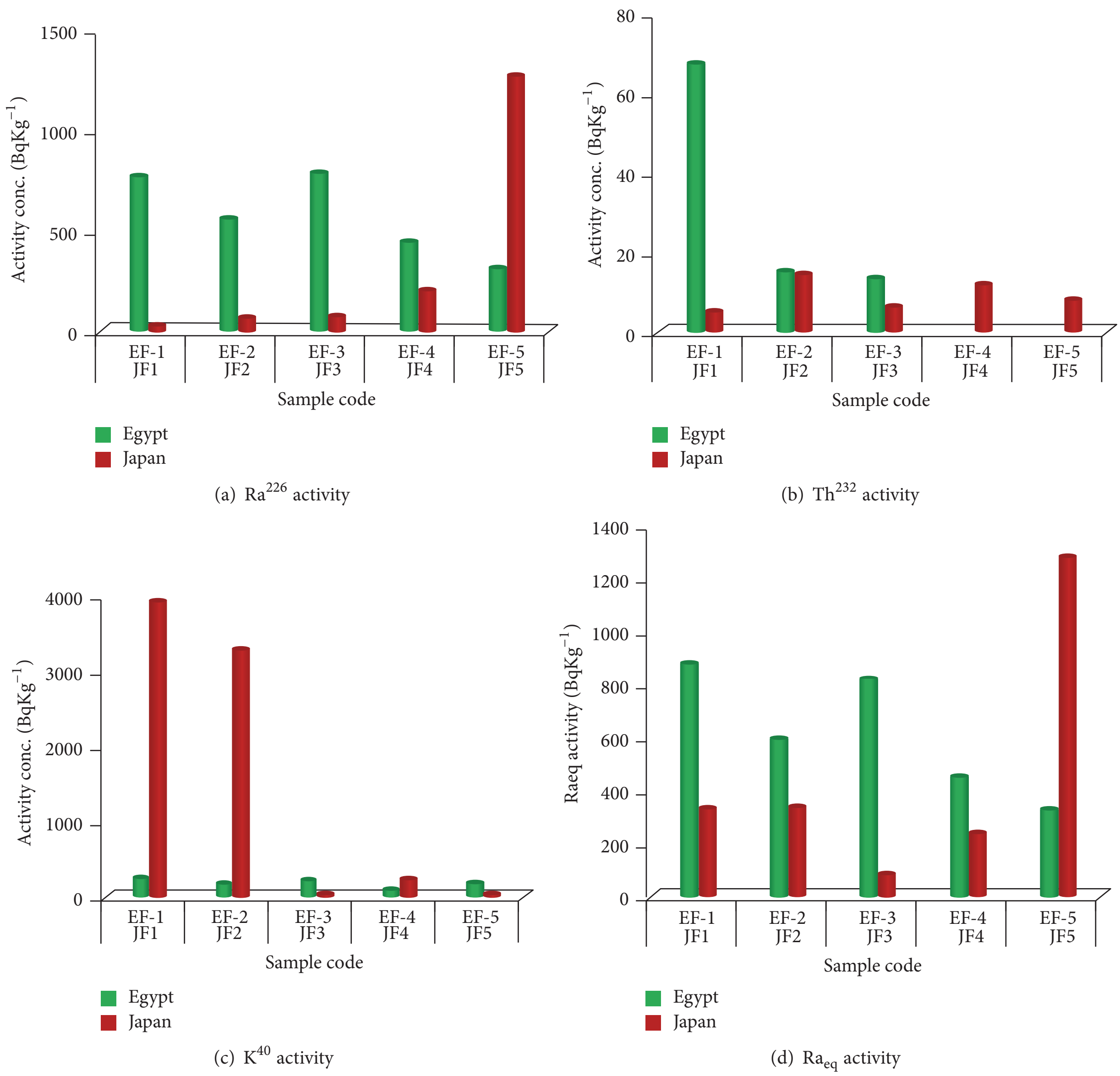


Figure 1: Comparison of natural radioactivities and radium equivalent of Egyptian and Japanese fertilizers.

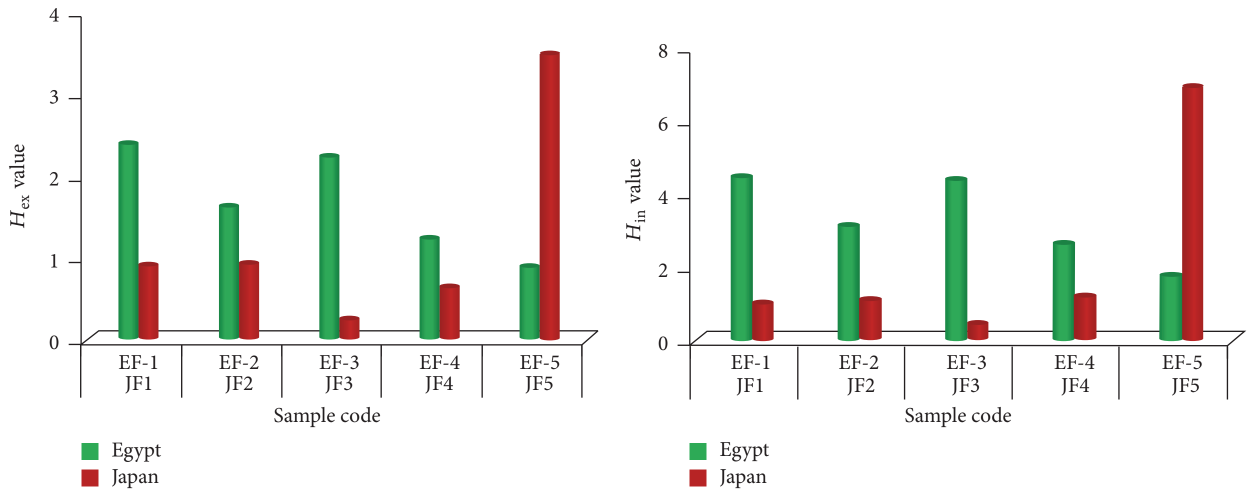
3. Results and Discussion

The specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in commonly used fertilizer samples in Egypt and Japan are given in Tables 1 and 2. The respective average radionuclide activities of ^{226}Ra, ^{232}Th, and ^{40}K in the Egyptian fertilizer samples were $571 \pm 20\,\mathrm{Bq\,kg^{-1}}$, $19 \pm 5\,\mathrm{Bq\,kg^{-1}}$, and $182 \pm 40\,\mathrm{Bq\,kg^{-1}}$ while, for the Japanese fertilizers, they were $325 \pm 2\,\mathrm{Bq\,kg^{-1}}$, $9 \pm 1\,\mathrm{Bq\,kg^{-1}}$, and $1500 \pm 10\,\mathrm{Bq\,kg^{-1}}$, as shown in Figure 1 and Tables 1 and 2. The radionuclide concentrations in Japanese fertilizer were less than those of Egyptian fertilizers except for potassium, as seen in Figure 1. The radionuclide concentration of ^{40}K is much higher in Japanese fertilizer samples and especially sample JF-1. Both Egyptian and Japanese fertilizers maintain radionuclide concentrations less than the recommended limits by UNSCEAR, 2008, [22].

To compare the radiation effect of different radionuclides in a sample UNSCEAR [22, 23] has introduced the radium equivalent concentration ($\mathrm{Ra_{eq}}$).

$$\mathrm{Ra_{eq}} = A_{\mathrm{Ra}} + 1.43 A_{\mathrm{Th}} + 0.077 A_{\mathrm{K}}, \qquad (2)$$

where A_{Ra}, A_{Th}, and A_{K} are activities of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in $\mathrm{Bq\,kg^{-1}}$. Radium equivalent concentration was calculated based on the estimation that $370\,\mathrm{Bq\,kg^{-1}}$ of ^{226}Ra, $259\,\mathrm{Bq\,kg^{-1}}$ of ^{232}Th, and $4810\,\mathrm{Bq\,kg^{-1}}$ of ^{40}K produce the same equivalent γ-ray dose. The value of $\mathrm{Ra_{eq}}$ of fertilizer must be less than $370\,\mathrm{Bq\,kg^{-1}}$ to keep γ-ray dose below $1.5\,\mathrm{mSv\,y^{-1}}$. The average radium equivalent concentration in Egyptian fertilizer was $613 \pm 33\,\mathrm{Bq\,kg^{-1}}$ while for Japanese fertilizer, it was $454 \pm 5\,\mathrm{Bq\,kg^{-1}}$, as shown in Tables 3 and 4. All Japanese fertilizers have radium equivalent concentrations



(a) External hazard index (b) Internal hazard index

FIGURE 2: Comparison of external and internal indexes of Egyptian and Japanese fertilizers.

TABLE 3: The Radium equivalent and hazard index in Egyptian fertilizers.

Sample code	Radium equivalent Ra_{eq}, ($Bq\,kg^{-1}$)	External hazard index (H_{ex})	Internal hazard index (H_{in})
EF-1	877 ± 57	2.37 ± 0.08	4.43 ± 0.24
EF-2	592 ± 43	1.60 ± 0.05	3.11 ± 0.17
EF-3	819 ± 37	2.21 ± 0.06	4.33 ± 0.17
EF-4	449 ± 13	1.21 ± 0.03	2.41 ± 0.07
EF-5	326 ± 16	0.88 ± 0.01	1.72 ± 0.08

less than the recommended limit except JF5 ($1277 \pm 8\,Bq\,kg^{-1}$) while all Egyptian fertilizer samples have radium equivalent concentrations greater than recommended limit except EF5 ($326 \pm 16\,Bq\,kg^{-1}$), as seen in Figure 1.

The external hazard index (H_{ex}) was determined from (3) [24, 25]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}, \tag{3}$$

where A_{Ra}, A_{Th}, and A_K are the activities of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in $Bq\,kg^{-1}$. The external hazard index should be less than unity in order to keep γ-radiation dose less than $1.5\,mSv\,y^{-1}$. The calculated external hazard index for Egyptian fertilizers had an average of 1.65 ± 0.04 (Table 3) while it was 1.22 ± 0.06 for Japanese fertilizer samples (Table 4). The external hazard index for Japanese fertilizers was less than the recommended limit except for the sample of JF5 (3.45 ± 0.09) while its value was higher than the recommended limit for Egyptian fertilizers except the one sample of EF-5 (0.88 ± 0.01), as shown in Figure 2(a). In addition to the external hazard, radon and its short-lived products are also hazardous to respiratory organs. The internal exposure to radon and its progeny produced is quantified by an internal hazard index (H_{in}) which can be defined as [24, 25]

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}. \tag{4}$$

For the safe use of a material, H_{in} should be less than unity. The average value of the internal hazard index was 3.20 ± 0.15 (Table 3 and Figure 2(b)) for Egyptian fertilizers, all of which are higher than the recommended limit. This means that the use of Egyptian fertilizers should be subject to precautions. On the other hand, Japanese fertilizers had a mean internal hazard index of 2.11 ± 0.13, with JF-1 and JF-3 having internal hazard index less than unity (Table 4 and Figure 2(b)).

The use of fertilizers in agriculture is a possible source of exposure for the public. Elevated radionuclides exposure of the public might be expected, for example, to the workers in sites being developed for housing. [22]. The absorbed dose (D) due to γ-rays emitted at 1 m of air above ground can be calculated from the following equation [24]:

$$D\left(nGy\,h^{-1}\right) = 0.462 A_{Ra} + 0.621 A_{Th} + 0.042 A_K. \tag{5}$$

The absorbed dose was calculated for the samples as shown in Tables 5 and 6 and Figure 3(a). The radiation absorbed dose was varied from $141 \pm 7\,nGy\,h^{-1}$ (EF5) to $380 \pm 26\,nGy\,h^{-1}$ (EF1) with a mean value of $264 \pm 15\,nGy\,h^{-1}$ for Egyptian fertilizers. For Japanese fertilizers it varied from $38 \pm 1\,nGy\,h^{-1}$ (JF3) to $546 \pm 4\,nGy\,h^{-1}$ (JF5) with a mean value of $210 \pm 2\,nGy\,h^{-1}$. All Egyptian fertilizers had absorbed radiation dose values greater than the limit recommended by UNSCEAR [23], of $59\,nGy\,h^{-1}$ as also Japanese fertilizers except JF3, as given in Tables 5 and 6 and Figure 3(a). Therefore, except for one sample, all the Egyptian and Japanese fertilizers gave an absorbed dose larger than recommended so that these materials should be used with precautions.

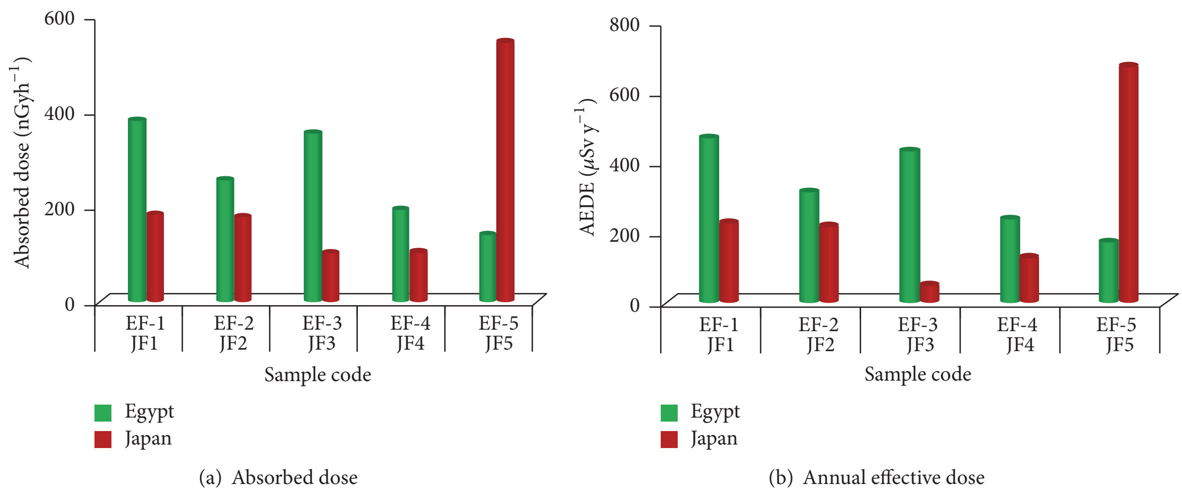


Figure 3: Comparison of absorbed and annual effective doses of Egyptian and Japanese fertilizers.

Table 4: The Radium equivalent and hazard index in Japanese fertilizers.

Sample code	Radium equivalent Ra_{eq}, (Bq kg^{-1})	External hazard index (H_{ex})	Internal hazard index (H_{in})
JF1	333 ± 5	0.90 ± 0.05	0.97 ± 0.10
JF2	336 ± 5	0.91 ± 0.08	1.07 ± 0.11
JF3	87 ± 2	0.23 ± 0.03	0.43 ± 0.06
JF4	235 ± 3	0.63 ± 0.04	1.18 ± 0.09
JF5	1277 ± 8	3.45 ± 0.09	6.87 ± 0.28

Table 5: Absorbed and annual effective dose and hazard indexes of fertilizers used in Egypt.

Sample code	Absorbed dose (nGy h^{-1})	Annual effective dose (μSv y^{-1})	Gamma index	Alpha index
EF-1	380 ± 26	468 ± 3	2.96 ± 0.20	3.81 ± 0.16
EF-2	255 ± 20	314 ± 2	1.99 ± 0.10	2.79 ± 0.09
EF-3	353 ± 17	434 ± 2	2.75 ± 0.13	3.91 ± 0.12
EF-4	193 ± 6	237 ± 1	1.50 ± 0.04	2.21 ± 0.06
EF-5	141 ± 7	173 ± 1	1.10 ± 0.06	1.56 ± 0.07

Table 6: Absorbed and annual effective dose and hazard indexes of fertilizers used in Japan.

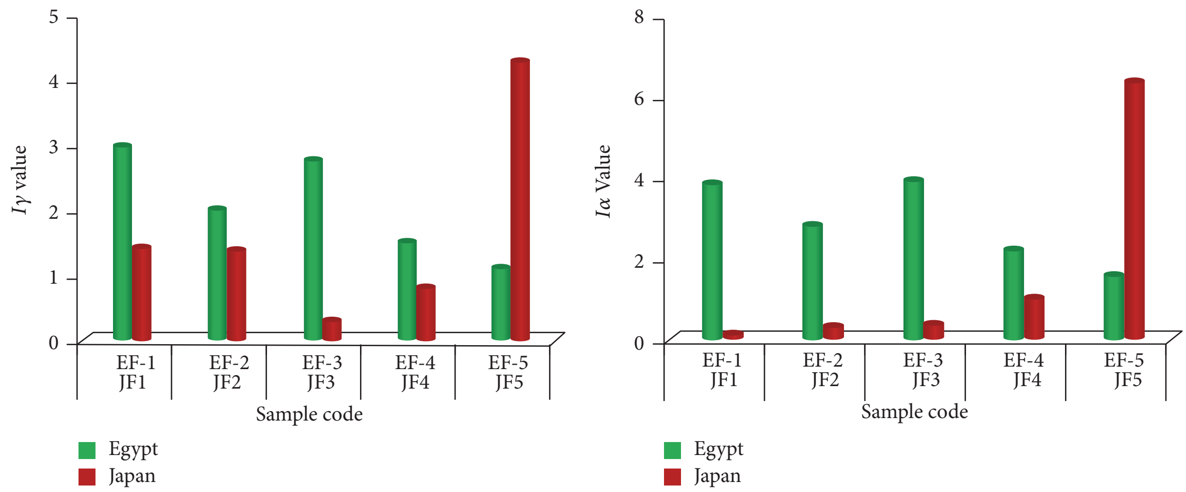
Sample code	Absorbed dose (nGy h^{-1})	Annual effective dose (μSv y^{-1})	Gamma index	Alpha index
JF1	182 ± 3	225 ± 3	1.41 ± 0.07	0.13 ± 0.03
JF2	177 ± 2	218 ± 3	1.37 ± 0.05	0.31 ± 0.05
JF3	38 ± 1	47 ± 1	0.29 ± 0.03	0.37 ± 0.07
JF4	103 ± 2	127 ± 2	0.80 ± 0.04	1.00 ± 0.11
JF5	546 ± 4	672 ± 4	4.26 ± 0.19	6.32 ± 0.25

The annual effective dose (E) from γ-rays emitted from ^{226}Ra, ^{232}Th and ^{40}k in the samples was calculated from [22]

$$E = D\left(\text{nGy h}^{-1}\right) \times 8760\left(\text{h y}^{-1}\right) \times O \times C\left(\text{mSv/nGy}\right), \qquad (6)$$

where O is the occupancy factor and C is the absorbed to effective dose conversion factor of 0.7×10^{-6} Sv per Gy. The annual effective doses from γ-rays emitted by ^{226}Ra, ^{232}Th and ^{40}k in the samples varied from $173 \pm 1\ \mu$Sv y^{-1} (EF5) to $468 \pm 3\ \mu$Sv y^{-1} (EF1) with a mean value of $325 \pm 2\ \mu$Sv y^{-1} for the Egyptian samples. For Japanese samples they varied from $47 \pm 1\ \mu$Sv y^{-1} (JF3) to $672 \pm 4\ \mu$Sv y^{-1} (JF5) with a mean value of $258 \pm 3\ \mu$Sv y^{-1}, as shown in Tables 5 and 6 and Figure 3(b). The annual effective dose of all studied samples of Egyptian and Japanese fertilizers is less than the recommended limiting value of 480 μSv y^{-1} [22] except for the sample JF5.

The γ-ray radiation hazards associated with the natural radionuclides in fertilizer materials can be assessed by means of the radioactivity level index, I_γ. According to the European



(a) Gamma index

(b) Alpha index

Figure 4: Comparison of gamma and alpha indexes of Egyptian and Japanese fertilizers.

Table 7: Comparison of radiological indexes in the present work and their values in literature.

Country	Sample	Radium equivalent (Bq/kg)	Gamma index (I_γ)	References
Egypt	Fertilizer	613 ± 33	2.06 ± 0.11	Present work
Japan	Fertilizer	454 ± 5	1.63 ± 0.08	Present work
Algeria	NPK fertilizer	1168	9.6	[3]
Brazil	NPK fertilizer	1772	12.3	[11]
Saudi Arabia	NPK fertilizer	275	—	[12]
Egypt (Qena)	Phosphate fertilizer	462	3.1	[13]
Bangladesh	Triple superphosphate	374	—	[14]

Commission guidelines, I_γ should be less than 6 $(2 < I_\gamma < 6)$ for a radiation dose of $1\,\mathrm{mSv\,y^{-1}}$ [26]. The γ-ray index (I_γ) can be calculated from [26]

$$I_\gamma = \frac{C_{\mathrm{Ra}}}{300} + \frac{C_{\mathrm{Th}}}{200} + \frac{C_{\mathrm{K}}}{3000}. \quad (7)$$

I_γ of the Egyptian samples varied from 1.10 ± 0.06 for (EF-5) to 2.96 ± 0.20 for (EF-1) with a mean value of 2.06 ± 0.11. For the Japanese samples, it varied from 0.29 ± 0.03 for (JF3) to 4.26 ± 0.19 for (JF5) with a mean value of 1.63 ± 0.08, as can be seen in Tables 5 and 6 and Figure 4(a). All the measured samples had a radioactivity level index less than 6, so any of these samples can be used without special precautions [26].

Alpha radiation due to the released radon from samples is called alpha index (I_α) which can be calculated from (8), [26]. Alpha index should be less than unity to reflect a radium concentration value less than $200\,\mathrm{Bq\,kg^{-1}}$ (the upper recommended value) which leads to a released radon concentration less than $200\,\mathrm{Bq\,m^{-3}}$.

$$I_\alpha = \frac{A_{\mathrm{Ra}}}{200}. \quad (8)$$

Alpha index of the Egyptian samples varied from 1.56 ± 0.07 (EF5) to 3.91 ± 0.12 (EF3) with a mean value of 2.86 ± 0.10. For Japanese samples, it varied from 0.13 ± 0.03 (JF1) to 6.32 ± 0.25 (JF5) with a mean value of 1.63 ± 0.11, as seen in Tables 5 and 6 and Figure 4(b). The values of alpha index for Egyptian fertilizers were more than unity while, for Japanese samples, it was less than unity except a sample of JF5.

All these radiological indexes were actually initially developed and established for construction materials but they were used during this study and other previous studies in literature [11–14] to show how much the workers and public could receive radiation dose from fertilizer materials. Table 7 shows a comparison of the estimated radiological indexes values during this work and their values in previous studies in literature. Radium equivalent in the present work was less than its value for fertilizer used in Algeria and Brazil but it was greater than its value for fertilizer used in Saudi Arabia and Bangladesh. Gamma index behaved the trend as radium equivalent, as seen in Table 7.

4. Conclusion

Natural radioactivity of ^{226}Ra, ^{232}Th, and ^{40}K in different types of fertilizers used in Egypt and in Japan was measured using a high purity germanium detector. The specific activities in Egyptian samples ranged from 312 ± 14 to 782 ± 24 $\mathrm{Bq\,kg^{-1}}$ for ^{226}Ra, ND to 67 ± 13 $\mathrm{Bq\,kg^{-1}}$ for ^{232}Th, and 88 ± 22 to 251 ± 94 $\mathrm{Bq\,kg^{-1}}$ for ^{40}K. In Japanese samples these specific activities ranged from 25 ± 1 to $1265 \pm 5\,\mathrm{Bq\,kg^{-1}}$ for ^{226}Ra, 5 ± 2 to $15 \pm 1\,\mathrm{Bq\,kg^{-1}}$ for ^{232}Th, and 31 ± 5 to $3909 \pm 21\,\mathrm{Bq\,kg^{-1}}$ for ^{40}K. The specific radioactivities of Egyptian fertilizers are much higher than their values for Japanese fertilizers but still in the range of recommended limits of UNSCEAR 2008. The radiological hazard indexes, of radium equivalent activities (Ra_{eq}), external and internal indexes, gamma index, absorbed radiation, and annual effective doses of the Egyptian fertilizers were higher than the values for Japanese fertilizers and higher than the respective safely values of $370\,\mathrm{Bq\,kg^{-1}}$, unity, $59\,\mathrm{nGy\,h^{-1}}$, and $480\,\mu\mathrm{Sv\,y^{-1}}$. From all of these results, we deduce that the amount of fertilizers that showed high radioactivities should be decreased and used with precautions.

Conflicts of Interest

The authors declared that there are no conflicts of interest regarding the publication of this paper.

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