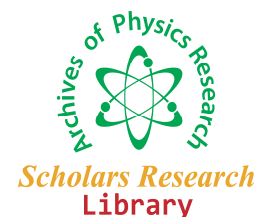




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Natural radioactivity of some local and imported fertilizers in Basrah Governorate/Iraq

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ABSTRACT

The naturally occurring activity concentration of radionuclides ^{226}Ra , ^{232}Th , ^{228}Ra , ^{238}U and ^{40}K has been measured in different types of fertilizer samples used in Basrah Governorate using sodium iodide NaI(Tl) gamma spectrometry. The results of measurements showed that the mean value and range of specific activities for ^{226}Ra , ^{232}Th and ^{40}K activities in the nitrogen, phosphorus and potassium fertilizers are 107.0 ± 8.7 Bq/kg (8.6 - 410), 108.0 ± 7.6 Bq/kg (4.1 - 397.5) and 1207.0 ± 9.8 Bq/kg (201.2 - 4237.7) Bq/kg, respectively. The maximum radium activity found in Crop Complex fertilizer with respect to organic fertilizers under investigation. Radium equivalent activity exceeds 370 Bq/kg, in some samples, the maximum permissible limit for radiation dose for all present samples. This study could be useful as baseline data for radiation exposure to fertilizers and their impact on human health.

Keywords: Natural radioactivity, Fertilizers, Gamma Spectroscopy, NaI(Tl)

INTRODUCTION

The exposure of the public to natural radioactivity has been estimated by the UNSCAR, which concluded an effective average annual dose equivalent to 2.4 mSv/y per individual [1-3]. Natural radioactivity arises mainly from the primordial radionuclides, such as ^{40}K , and the radionuclides from ^{238}U and ^{232}Th series and their decay products, which are present at trace levels in all ground formations [4]. This natural radioactivity in soil may vary considerably from one type to other. In soil, one source of radioactivity other than those of natural origin is mainly due to extensive use of fertilizer which is rich in phosphate and accordingly the radioactive ^{40}K used for agricultural purpose [5]. The knowledge of specific activities or concentrations and distributions of the radionuclides in these fertilizers are of interest since it provides useful information in the monitoring of environment radioactivity. Gamma radiation emitted from fertilizers represents one of the main external source of irradiation of the human body. The fertilizers added to soil in order to reach high agriculture productivity, for example adding the NPK to the soil replace the nutrients in soils [6]. Only nuclides with half-lives comparable with the age of the earth or their corresponding decay products, existing in terrestrial materials, such as ^{40}K , ^{238}U and ^{232}Th are of great interest. Abnormal occurrences of uranium and its decay products in ore rocks and fertilizer are the main sources of high natural background areas that have been identified in several areas of the world [7]. From the point of view of the international Atomic Energy Agency of radio nuclides in food and environment IAEA(8), it is necessary to estimate the dose limits of occupier exposures and to measure the natural environmental radiation level provided by ground, air, water, fertilizer etc. for estimation of the exposure to natural radiation sources. The UNCEAR, 1993[9] estimated the general range of ^{238}U is 37- 4900 Bq/kg and for ^{226}Ra a range of 100 - 10,000 Bq/kg in different

phosphate fertilizers. The low level gamma ray spectroscopy is suitable for both qualitative and quantitative determination of gamma ray emitting nuclide in the environment.

The aim of our study was to estimate the radioactivity concentration of commercially and locally available fertilizers and to draw a conclusion on the radiation exposure due to agricultural uses of chemical fertilizer in Basrah governorate in Iraq.

MATERIALS AND METHODS

Experimental Work

Sixteen fertilizer samples of 7 different brands were collected from local market and factory in Basrah governorate. All brand of fertilizers are solid and some of them need to dissolve in water before used (leaf fertilizers). After collection the sample were grinded to fine powder, heated in the oven at 100°C for 24h to remove moisture, sealed in plastic containers, and then stored for 30days to allow the equilibrium between ^{226}Ra and ^{222}Rn . The activity concentration of ^{226}Ra , ^{228}Ra , ^{238}U , ^{232}Th and ^{40}K was estimated from the gamma spectrum using Na(Tl) detector 3x3 inch with a 1024 channel computer analyzer USX supplied by Spectrum Technique Company. The detector was employed with lead shielding, 4 cm thickness, which reduced the background. The detector was calibrated using standard sources of ^{57}Co (peak 122 keV), ^{137}Cs (peak 662 keV) and ^{60}Co (peaks 1173, 1333 keV). The detector resolution is about 8% at 662 KeV of ^{137}Cs . The efficiency calibration was achieved using eight standard sources include the calibration sources. The samples were placed over the detector for at least 12h for counting. The system was running freely, for the same live time, to evaluate the background spectrum. One of the fertilizer spectrum, sample no 2, is shown in figure 1.

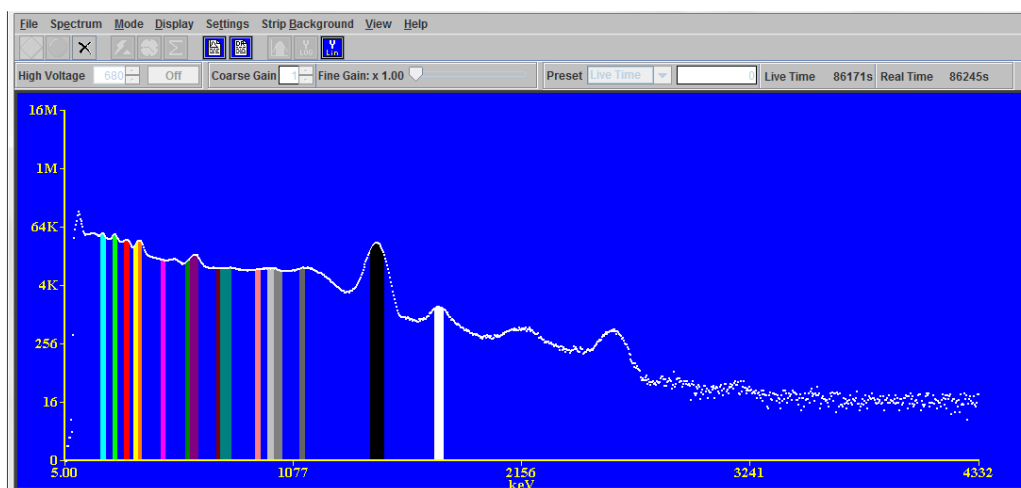


Figure 1. The NaI(Tl) spectrum of sample number 2, where most of the peaks are clearly visible. The color represents the area under the peak of interest

THEORETICAL ASPECT

After measuring the count rate (area under the peak) for each peak shown in figure 1, the activity concentration for each environmental isotopes calculated from [10]

$$A = \frac{\text{Net count}}{\epsilon \times I_\gamma \times M \times t} \quad (1)$$

where ϵ is absolute gamma peak efficiency of the detector at this particular gamma-ray energy, I_γ decay intensity for the specific energy peak (including the decay branching ratio information), M the mass of the sample in kg and t is the counting time of the measurement in second.

RADIUM EQUIVALENT ACTIVITY

Radium equivalent activity (Ra_{eq}) is used to assess the hazards associated with materials that contain ^{226}Ra , ^{232}Th and ^{40}K in Bq kg^{-1} , which is, determined by assuming that 370 Bq kg^{-1} of ^{226}Ra or 260 Bq kg^{-1} of ^{232}Th or 4810 Bq kg^{-1} of ^{40}K produce the same γ dose rate. The Ra_{eq} of a sample in (Bq kg^{-1}) can be achieved using the following relation [11];

$$Ra_{eq} = (A_{Ra}) + (A_{Th} \times 1.43) + (A_K \times 0.077) \quad (2)$$

The published maximal permissible Ra_{eq} is 370 Bq kg^{-1} [1].

EXTERNAL HAZARD INDEX

The external hazard index is an evaluation of the hazard of the natural gamma radiation. The prime objective of this index is to limit the radiation dose to the admissible permissible dose equivalent limit around 1mSv y^{-1} . In order to evaluate this index, one can use the following relation [11]

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_K/4810) \quad (3)$$

This model takes into consideration that the external hazard which is caused by gamma-rays corresponds to a maximum radium-equivalent activity of 370 Bq/kg for the soil.

THE ANNUAL EFFECTIVE DOSE RATE

In order to estimate the annual effective dose rate in air, the conversion coefficient from absorbed dose in air to effective dose received by an adult must be considered. This value is published in UNSCEAR 2000 and UNSCEAR 1993, to be 0.7 Sv Gy^{-1} for environmental exposure to gamma rays of moderate energy. The outdoor occupancy factor is about 0.2 [1]. The annual effective dose equivalent is given by the following equation [11];

$$AEDE_{oo}(\mu\text{Sv/y}) = D(n\text{Gy/h} \times 8760(h/y) \times 0.2 \times 0.7(\text{Sv/Gy}) \times 10^{-6} \quad (4)$$

$$\text{where } D\left(\frac{n\text{Gy}}{h}\right) = 0.0417A_K + 0.462A_{Ra} + 0.606A_{Th} \quad (5)$$

The world average annual effective dose equivalent (AEDE) from outdoor or indoor terrestrial gamma radiation only is 0.560 mSv/year [1].

RESULTS AND DISCUSSION

For calculation of the radioactivity concentration for each environmental (NORM), or (TENORM), one has to recognized the belong city of each peak according to gamma decay of each isotope [12]. For ^{226}Ra we are looking for the gamma ray lines 295 keV (19.2%), 352 keV (37.1%), 609 keV (46.1%), 1120 keV (15%) and 1760 keV (15.4%). The peak of 186 keV assumed to be from ^{235}U since it has slight effect on the total concentration after subtracting the background, 42.8% for Ra and the rest for ^{235}U . The determination of existence of ^{232}Th was achieved by 338 keV (12%), 911 keV (29%), 964 keV (5.05%) and 969 keV (17%). The case of ^{238}U is recognized by 1001 keV (83%), 766 keV (29%) and 2204 keV (5%). For ^{40}K , which directly determined using 1460 keV (10%) peak. Using all the information mentioned above one can calculate the activity concentration in the fertilizers used in the present work and tabulated the results in table 1,

The rest of the parameters are listed in table 2, and they are Ra_{eq} , H_{ex} , D and $AEDE_{oo}$. From table 1, one can recognize that maximum value of ^{226}Ra concentration is $410.4 \pm 19.5 \text{ Bq/kg}$ (sample 2, Crop Complex, Belgium) and minimum value is $8.6 \pm 1.8 \text{ Bq/kg}$ (sample 6, NPK, Belgium). The average value for all fertilizer samples tested was $107.0 \pm 8.7 \text{ Bq/kg}$. The average values of ^{232}Th , ^{228}Ra , ^{238}U and ^{40}K are $108.0 \pm 7.6 \text{ Bq/kg}$, $8.1 \pm 0.16 \text{ Bq/kg}$, $1.31 \pm 0.03 \text{ Bq/kg}$ and $1207.3 \pm 9.68 \text{ Bq/kg}$ respectively. El-Taher et al. [13], reported a table of comparison between the activity concentration of phosphate fertilizer in different countries ranging from 10, 2.8 Bq/kg to 5022, 394, 397 Bq/kg for ^{226}Ra , ^{232}Th and ^{40}K respectively. Wassila et al. [14], also reported a table for comparison of their measurement of fertilizers and soil radioactivity and different countries. The table shows ranges for ^{226}Ra , ^{232}Th and ^{40}K as 121 Bq/kg to 2150 Bq/kg , 26 Bq/kg to 753.9 Bq/kg and 4 Bq/kg to 11644 Bq/kg respectively. Our results in table 1, show that the concentration ranges of ^{226}Ra , ^{232}Th and ^{40}K are; 8.6 Bq/kg to 410 Bq/kg , 4.1 Bq/kg to 397

Bq/kg and 201 Bq/kg to 2373 Bq/kg respectively and these agree with the results of reference [14]. Figure 3, shows the correlation between radium-226 concentration and thorium-232, correlation between radium-226 and potassium-40 and correlations between thorium-232 and potassium-40

Table1. The activity concentration of Radium, Thorium, Uranium and Potassium in fertilizer samples

S. No.	Sample ID	^{226}Ra Bq/kg	^{232}Th Bq/kg	^{228}Ra Bq/kg	^{238}U Bq/kg	^{40}K Bq/kg
1	NPK, Switzerland	45.1±10.4	35.5±2.5	0.01±0.01	0.01±0.01	2373.4±14.1
2	Crop Complex, Belgium	410.4±19.5	255.3±17.9	0.01±0.01	0.01±0.01	2137.3±10.7
3	NPK, Belgium	36.3±8.1	52.4±3.2	0.93±0.01	0.21±0.01	4103.6±19.2
4	Urea-Calacum phosphate, Belgium	65.4±11.6	80.5±5.6	1.1±0.01	0.3±0.01	201.2±9.2
5	Calacum Nitra, Belgium	190.1±5.2	52.3±3.7	4.0±0.1	1.0±0.01	228.6±1.2
6	NPK, Belgium	8.6±1.8	42.5±3.0	0.01±0.01	0.30±0.01	1608.8±10.0
7	NPK, Belgium	45.5±10.0	285.2±20.5	53.47±1.0	0.0	286.3±1.7
8	Potassium sulphate, Belgium	60.0±2.9	38.5±2.3	5.9±0.1	0.01±0.01	4237.7±18.1
9	Urea, Iran	53.6±8.6	4.1±0.3	0.01±0.01	0.01±0.01	270.7±2.1
10	Urea, Iraq	14.8±2.4	10.7±0.8	1.6±0.01	0.01±0.01	266.4±15.0
11	Super phosphate, Iraq	244.4±10.0	91.2±6.5	22.4±0.1	8.7±0.1	224.2±1.4
12	NPK –Dap, Jordan	164.8±14.4	74.7±5.3	8.0±0.1	7.2±0.1	239.7±1.4
13	Humi-plant, China	190.2±12.3	397.5±28.5	5.1±0.1	0.01±0.01	1124.9±6.7
14	NPK, China	107.8±8.1	263.1±18.6	23.6±1.0	1.4±0.1	534.8±30.7
15	NPK/SCOTTS, European	37.9±9.2	18.6±1.1	0.01±0.01	0.5±0.1	889.5±5.3
16	Tobseen-im, Japan	37.6±4.7	30.9±2.5	3.9±0.1	0.01±0.01	589.7±8.1

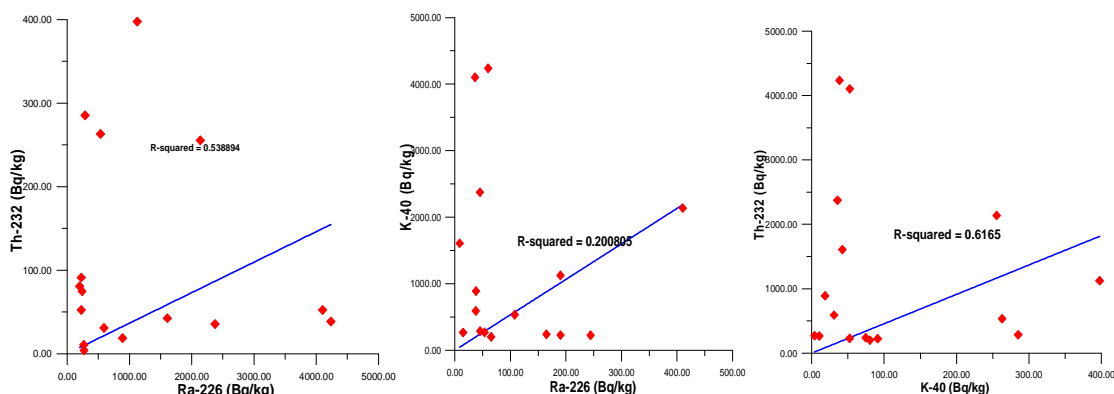


Figure 3. correlation between Ra-226 and Th232 on the left, between Ra-226 and K-40 in the middle and between Th-232 and K-40

Table 2. The Radium equivalent, External hazard and Annual Effective Dose, External, for different types of fertilizers

S. No	Sample ID	Ra_{eq}	H_{ex}	D nGy/h	$\text{AEDE}_{\text{ext}}(\text{mSv/y})$
1	NPK, Switzerland	279±27	0.74	143	0.175
2	Crop Complex, Belgium	940±48	2.42	426	0.521
3	NPK, Belgium	427±31	1.13	225	0.275
4	Urea-Calacum phosphate, Belgium	137±8	0.33	62	0.076
5	Calacum Nitra, Belgium	283±10	0.74	124	0.152
6	NPK, Belgium	193±14	0.50	99	0.121
7	NPK, Belgium	475±32	1.15	209	0.256
8	Potassium sulphate, Belgium	441±23	1.17	231	0.282
9	Urea, Iran	80±11	0.215	37	0.045
10	Urea, Iraq	48±5	0.12	23	0.028
11	Super phosphate, Iraq	392±18	1.01	171	0.209
12	NPK –Dap, Jordan	290±21	0.75	127	0.155
13	Humi-plant, China	845±47	2.09	377	0.460
14	NPK, China	535±31	1.32	238	0.291
15	NPK/SCOTTS, European	133±16	0.35	66	0.081
16	Tobseen-im, Japan	127±15	0.33	61	0.075

In table 2, the maximum Ra_{eq} is 940±48 Bq/kg, which is high in comparison with the recommended value of ICRP for soil. The minimum value is 48±5 Bq/kg and the average is 351.6±22.3 Bq/kg, which is acceptable. However, the ranges in references [13,14] for different countries are; 366 Bq/kg to 639 Bq/kg and 114 Bq/kg to 1772 Bq/kg

respectively. The external hazard H_{ex} ranged from 0.12 to 2.42 means that some fertilizer has external hazard more than unity, but still below action level [13,14]. The annual effective dose from fertilizers due to gamma ray emission range from 0.045 mSv/y to 0.521 mSv/y and average value is 0.200 mSv/y, which is below the ICRP value and less than the 2.5 mSv/y for the total radiation dose of NORM.

CONCLUSION

Samples of fertilizers in the studied region were measured for their natural radioactivity content using gamma spectroscopy, the results obtained concluded that the application of fertilizer in the soil to enhance the crop yield enhances the activity concentration, radium, thorium and potassium of specific categories of fertilizer e.g. Crop Complex, Belgium. While sublimate other type of fertilizer like NPK will keep these values below the recommended limits by the ICRP as the maximum annual effective dose to member of the public. In the case of local fertilizers which contain phosphate, the radioactivity differs from one country to another depending on the soil geology. Also the use of fertilizers in large extent increases radionuclide in soil especially potassium, which is always present high gamma activity in fertilizers. The data presented in this study will serve as a base line survey for developing radionuclide concentration in the area by adding the fertilizer. Further investigation is still needed to measure the radioactivity in all categories of fertilizer used in the country.

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