Journal of Nuclear and Radiation Physics, Vol. 3, No. 1, 2008, pp. 53-62

EVALUATION OF NATURAL RADIOACTIVITY IN A CULTIVATED AREA AROUND A FERTILIZER FACTORY

H. M. Diab[1], S. A. Nouh[2], A. Hamdy[2], S. A. EL-Fiki[2]

[1]*National Center for Nuclear safety and Radiation Control, Atomic Energy Authority*
[2]*Physics Department, Faculty of Science, Ain Shams University, Cairo, Egypt*
E-mail: hnndiab@yahoo.co.uk

Rec. 1/4/2008 *Accept. 30/5/2008*

Soil samples were collected from a cultivated area, where phosphate fertilizers produced by Abu-Zabal factory. Radioactivity concentration was measured using HPGe (high purity germanium detection system). Specific activity levels due to ^{226}Ra, ^{232}Th and ^{40}K were measured in 23 fertilized soil samples at different depth from zero to 25 cm with a step of 5 cm depth. The average activity concentration (Bqkg^{-1}) in the collected soil samples were found to be in the ranged from 6.0 ± 1.2 to 87.5 ± 4.5 Bqkg^{-1} with an average value of 31.12 ± 2.22 Bq.kg^{-1} for ^{226}Ra, the ^{232}Th specific activities ranged from 3.8 ± 1.2 to 14.2 ± 3.3 Bqkg^{-1} with an average value of 10.96 ± 1.89 Bqkg^{-1} and the ^{40}K specific activities ranged from 71.8 ± 24 to 543.2 ± 26.5 Bqkg^{-1} with an average value of 264.1 ± 11.94 Bqkg^{-1}. The absorbed dose in air at the cultivated farm was found to be 31 nGyh^{-1} within the range (9.7-57.8) nGyh^{-1} while it is in the order of the world average level (57nGyh^{-1}). The radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}) and the annual dose equivalent were also calculated and compared with the international recommended values and found to be within the international level.

***Key words*: **Radioactivity concentration, pollution, monitoring, phosphate fertilizers

INTRODUCTION

Natural environmental radioactivity and the associated external exposure due to gamma radiation depend mainly on the geological and geographical conditions, and appear at different levels in the soils of each region in the world [1]. The specific levels of terrestrial environmental radiation are related to the composition of each lithologically separated area, and to the content of the rock from which the soils originate [2]. A number of human activities contribute to our natural radiation environment, and a number of non-nuclear industries will be due to their activities, “move” and possibly also further concentrate of some natural radioactive substances that can be found in the earth’s crust [3]. The environmental impact of fertilizer production depends on the raw materials, production processes and the status of the pollution control equipment [4]. In addition, fertilizer plants cause environmental harm through emissions of process specific chemicals into the air, discharges into water, and storage and solid waste problems. Actions should be undertaken to minimize the emissions and to clean up spills and solid wastes [5].

The phosphate material is very insoluble, and therefore, in its original state is practically unavailable as a plant phosphorus source [6]. Exposure of workers and the public to radiation from phosphate rock and fertilizer is therefore not unlikely. The European Commission has issued a draft proposal for revision of the Basic Safety Standards for the protection of workers and the general public against the dangers of ionizing radiation [7]. Phosphate materials used for production of phosphate fertilizers contain a minor quantity of radioactive material, mainly various members of the uranium and thorium series, and radioactive potassium. The fertilizer factory located at Abou-Zabal region, eastern Egypt, produces Calcium Di-phosphate, Sulfuric acid and phosphoric acid [7].

This study aims to assess the impact of phosphate industry which may represent a site of significant environmental contamination due to fertilizer production. The natural radioactivity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the collected samples were measured by the HpGe detection system around this fertilizer factory.

MATERIALS AND METHODS OF CALCULATION

Sampling and Sample Preparation

The phosphate fertilizer factory is located in Abou-Zabal region between 31^{o}36' - 31^{o}38'N and 30^{o}26 - 30^{o}28'E. The factory is 30 km to the northeast far from Cairo. The factory is surrounded by farmlands, owned by farmers. Ismailia side River (a branch of the River Nile) is located in front of the factory and it is the main source of land irrigation. Most of the cultivated area considered the main source of food for the people living in Cairo.

Soil sampling was carried out in the months of March–June 2005 and August 2006. Sampling was done using the Standard Sampling Methods [8]. The area was divided into 25 sites. The sampling sites are shown in figure 1. From each site, 5 samples were taken at a step of 5 cm depth covering 25 cm^2. The soil textures for all of the samples were very similar. The soil is slightly alkaline (PH 7.5), calcareous ($CaCO_3$ equivalent - 1.75 %), and low in organic matter (0.75%).

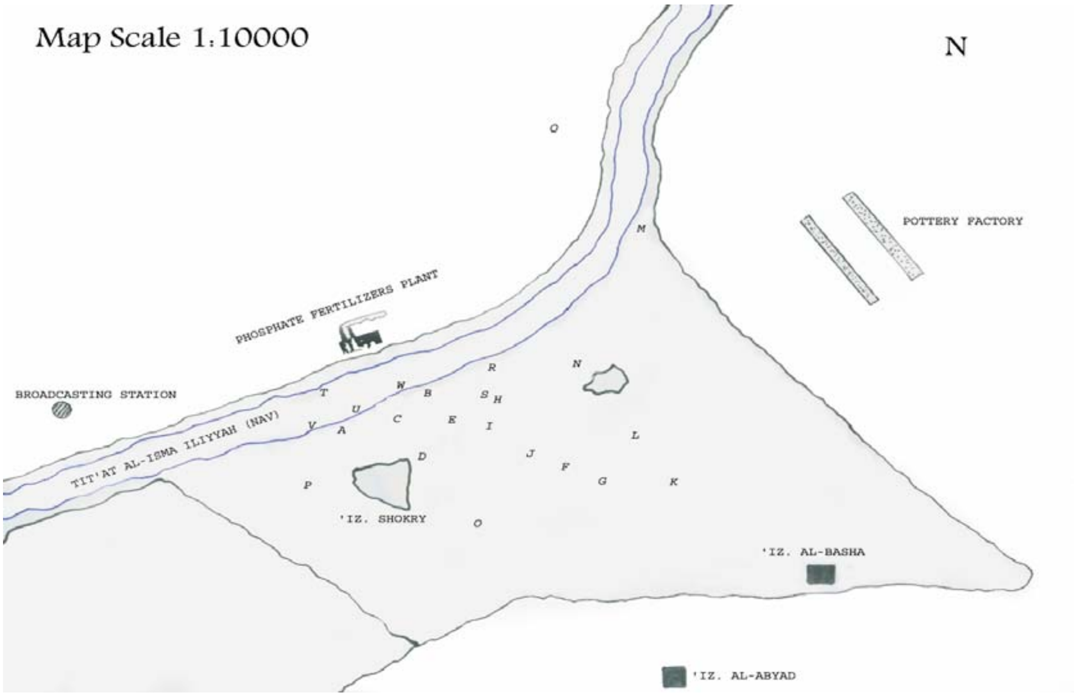

Figure 1. Location of the samples collected from the area in front of the factory

Over the selected area, some shore sediment and water samples were collected from the Ismailia side River.

The collected samples were transferred to labeled polyethylene bags, closed and transferred to the laboratory for preparation and measurement. The soil samples were prepared for analysis by drying, sieving and kept moisture free, by keeping over night in the electric oven at 115^{o}C. The samples were mechanically crushed and sieved through 0.8 mm mesh sieve. The sieved portion of the sample was transferred into a 100 ml Marinelli beaker for gamma spectrometry and sealed for four weeks to reach secular equilibrium between the thorium and radium contents of the sample and their daughters [8].

Instruments

A HpGe gamma-spectrometer with 40% efficiency and 2.0 keV resolution at 1.33 MeV photons of ^{60}Co, shielded by 4″ Pb 1 mm Cd and 1 mm Cu linked up to a multichannel analyzer was used for gamma measurements. The system was calibrated and the calibration quality control was carried out by using soil standard reference materials (IAEA-226 and IAEA-375) whose concentration of natural radioactivity has been certified by the IAEA.

Radiation hazard indices

To represent the activity levels of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them, a common radiological index has been introduced. This index is called radium equivalent (Ra_{eq}) activity and is mathematically defined by [1]:

$$Ra_{eq}(Bq.kg^{-1}) = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K\ ,$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. In the above relation, it has been assumed that 10 Bq kg^{-1} of ^{226}Ra, 7 $Bq.kg^{-1}$ of ^{232}Th and 130 $Bq.kg^{-1}$ of ^{40}K produce equal gamma dose. The absorbed dose rates (*D*) due to gamma radiations in air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on guidelines provided by UNSCEAR 2000 [1]. We assumed that the contributions from other naturally occurring radionuclides were insignificant. Therefore, *D* can be calculated according to UNSCEAR 2000.

$$D(nGy.h^{-1}) = 0.462\ A_{Ra} + 0.621\ A_{Th} + 0.0417\ A_K$$

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose ($0.7Sv.Gy^{-1}$) and outdoor occupancy factor (0.2) proposed by UNSCEAR 2000 [1] are used. Therefore, the annual effective dose rate ($mSv.yr^{-1}$) was calculated by the following formula [1]:

$$\text{Effective dose rate } (mSv.yr^{-1}) = D\ (nGy.h^{-1}) \times 8760\ h.yr^{-1} \times 0.7 \times (10^3\ mSv\ /10^9)\ nGy \times 0.2$$
$$= D \times 1.21 \times 10^{-3}\ \ (mSv.yr^{-1})$$

A widely used hazard index (reflecting the external exposure) called the external hazard index H_{ex} is defined as follows [1]:

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

In addition to external hazard index, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index H_{in}, which is given by the equation

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810$$

The values of the indeces (H_{ex}, H_{in}) must be less than unity for the radiation hazard to be negligible.

EXPERIMENTAL RESULTS AND DISCUSSION

From the gamma spectrometric analysis, three naturally occurring radionuclides were determined ^{226}Ra, ^{232}Th and ^{40}K. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the raw material in the used fertilizer were found to be 1180.6 ± 0.3 $Bq.kg^{-1}$, 16.0 ± 20.1 $Bq.kg^{-1}$ and 1582.0 ± 0.67 $Bq.kg^{-1}$, respectively. The average activity concentration in the collected soil samples ranged from 6.0 ± 1.2 to 87.5 ± 4.5 $Bq.kg^{-1}$ with an average value of 31.12 ± 2.2 $Bq.kg^{-1}$ for ^{226}Ra. The ^{232}Th specific activities ranged from 3.8 ± 1.2 to 14.2 ± 3.3 $Bq.kg^{-1}$ with an average value of 10.96 ± 1.89 $Bqkg^{-1}$. The ^{40}K specific activities ranged from 71.8 ± 24 to 543.2 ± 26.5 $Bq.kg^{-1}$ with an average value of 264.1 ± 11.9 $Bq.kg^{-1}$. The average activity value of ^{226}Ra is about two times higher than that of ^{232}Th. The radionuclides detected in soil were shown in Table 1. Figures 2, 3, and 4 show the iso-specific activity maps (contour-radiological maps) expressed in $Bqkg^{-1}$, dry weight.

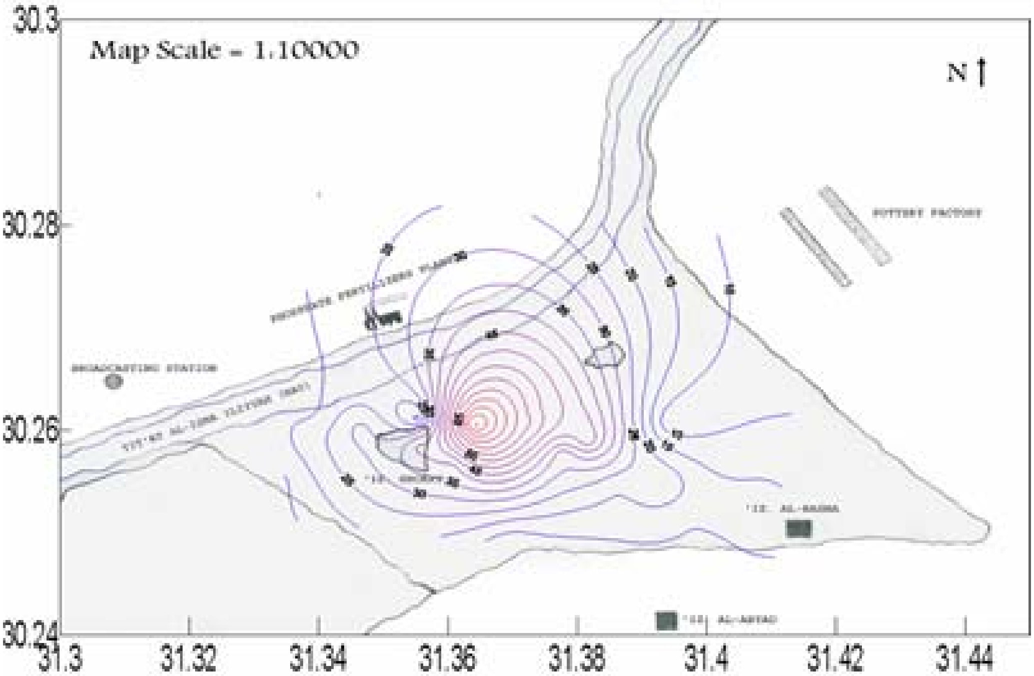

Figure 2. Contour diagram for the activity concentration of ^{226}Ra [$Bqkg^{-1}$] for the collected samples

These data show that, the activity concentration of naturally occurring radionuclides in soil samples were within the world average range except for the samples collected close to the stack. This indicates that the higher concentration levels are mainly due to fallout of phosphate dust generated during loading and processing of phosphate ore inside the phosphate fertilizers plant, which obvious increases the atmospheric release of the particles into the area affected by this industry.

The activity levels were found to follow lognormal distribution. As regard of activity with respect to depth, no correlation was found between depth and measured radioactivity showing that natural radioactivity is randomly distributed in different depths of the soil under investigation.

Table 1. Specific activities (Bq kg^{-1}) of ^{238}U (^{226}Ra), ^{232}Th and ^{40}K in soil samples

SAMPLE Code	Site	Ra-226	Th-232	K-40
S1	A	24.9±2.7	11.1±2.4	208.7±10.2
S2	B	87.5±4.5	12.9±3.5	231.2±10.5
S3	C	40.6±1.5	5.1±1.2	153±5.8
S4	D	39.7±2.1	10.9±1.7	226.8±16.1
S5	E	58.1±4.4	12.9±3.2	251.7±10.4
S6	F	25.1±1.4	13.5±1.5	543.2±26.5
S7	G	15.6±1.7	10.7±1.5	204.3±5.7
S8	H	49.2±2.4	13.6±1.8	217.4±7.2
S9	I	31.6±1.8	19.3±2.0	781.8±37.0
S10	J	20.7±2.5	14.2±3.3	207.6±13.1
S11	K	16.2±1.4	10.2±1.2	220.5±4.8
S12	L	16.3±0.9	9.5±0.7	203.5±2.6
S13	M	7.9±1.8	3.8±1.2	91.3±4.1
S14	N	6.0±1.2	4±2.2	118.8±14.7
S15	O	10.5±3.1	8.3±2.2	210.0±14.1
S16	P	14.3±3	12.4±2	218.0±8.2
S17	Q	12.0±0.9	7.8±0.7	143.3±2.7
S18	R	56.8±2.4	14.1±2.1	269.0±7.4
S19	S	58.3±2.5	14.0±1.5	517.0±25.7
SD1	T	18.3±1.5	6.8±3.6	390.0±17.1
SD2	U	31.7±3.2	10.0±1.6	71.8±24.0
SD3	V	22.2±2.1	5.5±1.1	550.0±10.5
SD4	W	12.6±1.33	6.28±2.6	210.3±3.1
Raw material	Stack	1180.6±0.3	16±20.1	1582.0±0. 7

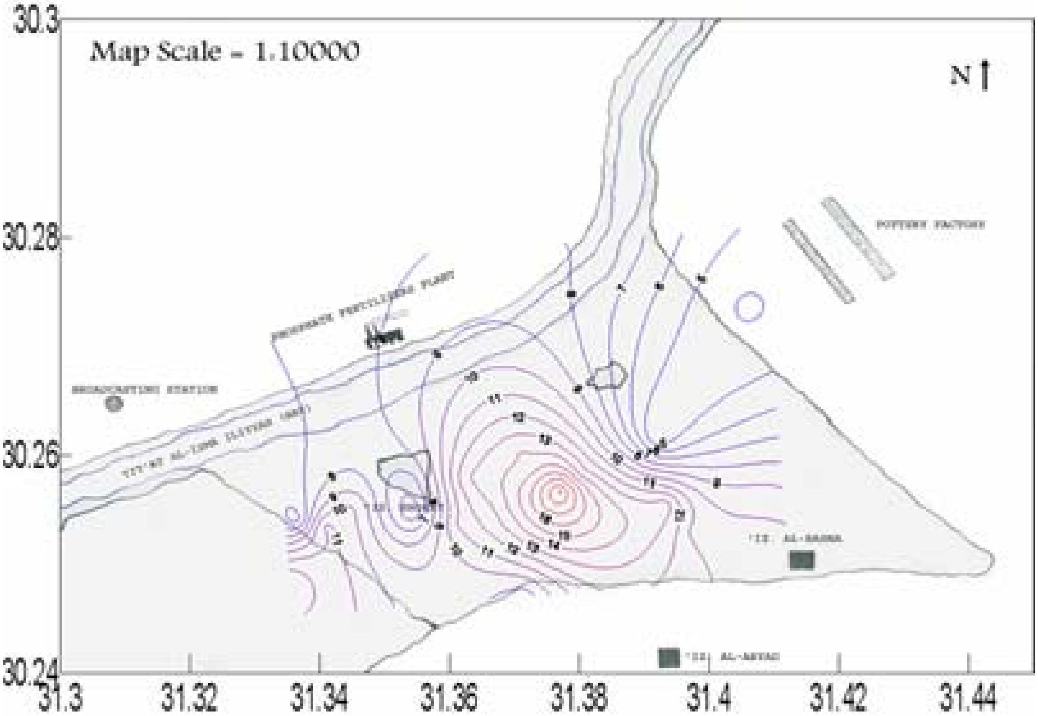

Figure 3. Contour diagram for the activity concentration of ^{232}Th [$Bqkg^{-1}$] for the collected Samples

The activity concentrations of the four shore sediments collected from Al-Ismailia lack were ranged from 12.6±1.3 to 31.7±3.2 $Bq.kg^{-1}$ for ^{226}Ra, from 5.5±1.1 to 10.0±1.6 $Bqkg^{-1}$ for ^{232}Th and from 71.8±24 to 550.0±10.5 $Bq.kg^{-1}$ for ^{40}K, and were found to be in good agreement with the average world level.

The gamma dose rates due to naturally occurring terrestrial radionuclides ^{226}Ra, ^{232}Th and ^{40}K were calculated based on their activities in soil samples, determined by gamma-ray spectrometry. The absorbed gamma dose rate due to these radionuclides varied from 9.8 to 58.9 $nGyh^{-1}$ with average 31.1$nGyh^{-1}$ which is within the value given in UNSCEAR report 2000 [57 (18-93) $nGyh^{-1}$]. The estimated annual mean effective dose comes out to be 0.2 $mSvy^{-1}$.

The external hazard index H_{ex} was calculated. It ranged from 0.055 to 0.33, with an average value of 0.17. The internal hazard index H_{in} was also calculated, the values of H_{in} ranged from 0.072 to 0.570, with an average value of 0.255. The total hazard index was ranged from 0.12 to 0.9 with average 0.8. The values of H_{ex} and H_{in} of all samples studied in this work are less than unity.

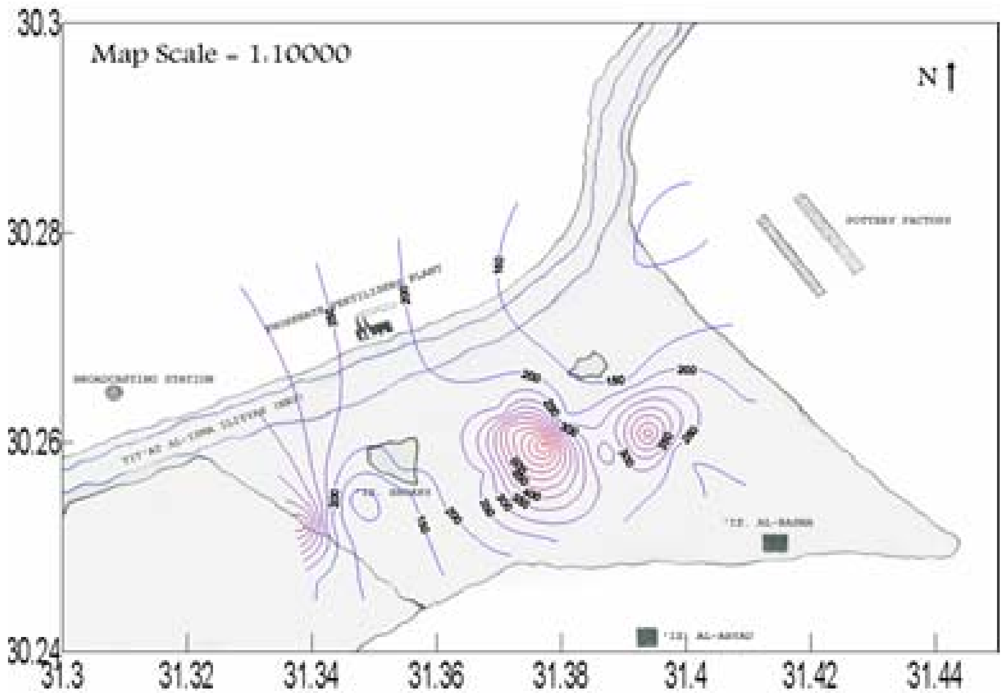

Figure 4. Contour diagram for the activity concentration of ^{40}K [$Bqkg^{-1}$] for the collected Samples

Phosphate rock is an important raw material used for manufacturing different types of phosphatic fertilizers. Phosphatic rocks are used as raw material in the formation of fertilizers. These fertilizers are the main source of radioactivity in cultivated soils [10]. The operation of phosphate fertilizer factories clearly enhanced the natural radiation levels of its close environment. The radioactivity concentration of $^{238}U(^{226}Ra)$, ^{232}Th and ^{40}K in soil samples which were measured in the Abou-Zabal region (south middle delta) which is very close to the studied area was ranged from 9.6 to 14.0 $Bqkg^{-1}$, 7.7 to 13.8 $Bqkg^{-1}$ and 99.6 to 140.1 $Bqkg^{-1}$, respectively [11]. In general, the radionuclides were present in farm soil and Nile delta soil were 31.12 ± 2.22 $Bqkg^{-1}$ for ^{226}Ra, 10.96 ± 1.89 $Bqkg^{-1}$ for ^{232}Th and 264.1 ± 11.94 $Bqkg^{-1}$ ^{40}K [12].

CONCLUSION

Because of the radionuclide activity concentrations in most of the process materials are slightly above the natural levels in soil, the need for specific measures to control radiological hazards to individuals and the environment is very limited. In most cases, normal occupational health and environmental protection measures designed for non-radiological hazards will be sufficient to protect against radiological hazards as well.

REFERENCES

[1] UNSCEAR "Sources, effects and risks of ionization radiation", United Nations Scientific Committee on the Effects of Atomic Radiation, *Report to the General assembly, with Annexes*, New York (2000).

[2] Habshi, F., "The recovery of uranium from phosphate rock: Progress and problems", *Proceeding of the 2nd International Congress on Phosphorus Compounds*, Institute Mondial du phosphate, Paris, 1980, pp 629-660.

[3] Scholten, L.C. and Timmermans, C.W.M., "Natural Radioactivity in Phosphate Fertilizer", *Nutrient Cycling Agrecosystem* **43(1-3)**, 103-107(2005).

[4] Bhatti, T.M., and Malik, K.A., "Phosphate Fertilizers a Potential Source for Uranium Recovery as by-Product", *National Institute for Biotechnology & Genetic Engineering (NIBGE) Faisalabad, Technical Report* No **PAEC/NIBGE-2/1994**.

[5] UNSCEAR (1988)**,** "*Sources, Effects and Risks of Ionizing Radiation*", United Nation Scientific Committee on the Effects of Atomic Radiation. United Nations, New York, USA.

[6] IAEA (**1973**) "*Safe Handling of Radionuclides*", Safety Series No. 1, Vienna.

[7] EC (European Commission), 1997. Radiation Protection 88, "*Recommendations for the Implementation of Title VII of the European Basic Safety Standards Directive (BSS) Concerning Significant Increase in Exposure due to Natural Radiation Sources. Directorate-General Environment, Nuclear Safety and Civil Protection*". Official Publication of the European Communities.

[8] Khater, A.J., *Environ. Radioact.* **71**, 33–41(2004).

[9] Beretka, J., Mathew, P.J., "Natural Radioactivity of Australian Building Materials, Industrial Wastes and By-products**",** *Health Physics.* **48(1)**, 87-95(1985).

[10] Tufail M., Akhtar Nasim, Waqas M, "Radioactive Rock Phosphate: The Feed Stock of Phosphate Fertilizers used in Pakistan", *Health Physics* **90(4)**:361-370(2006).

[11] Diab H. M., "*Nuclear Safeguards through measurements of radioactivity of Nuclear Materials Around Nuclear Facilities*", Ph.D., Thesis, Physics Dept., Ain Shams Univeristy, 1999.

[12] Nour Khalifa Ahmed, and Abdel Gabar Mohamed El-Arabi, "Natural radioactivity in farm soil and phosphate fertilizer and its environmental implications in Qena governorate, Upper Egypt", *Journal of Environmental Radioactivity* **84(**1), 51-64(2005).

تقدير النشاط الأشعاعى الطبيعى فى مساحة مزروعة حول مصنع اسمدة

حنان محمد دياب[١]، سمير نوح[٢]، أحمد حمدى[٢]، سعاد الفقى[٢]

١- المركز *القومى للأمان النووى والرقابة* الأشعاعية- هيئة الطاقة الذرية
٢- قسم الفيزياء- كلية العلوم- جامعة عين شمس[٢]- القاهرة – جمهورية مصر العربية

جمعت عينات تربة من الأراضى المسمدة حول مصنع سماد الفوسفات فى ابو زعبل. وتم قياس تركيز النشاط الأشعاعى باستخدام كاشف جرمانيوم عالى النقاوة، وكما تم تعيين النشاط الأشعاعى النوعى العائد الى الراديوم-٢٢٦ والثوريوم-٢٣٢ والبوتاسيوم-٤٠ فى ٢٣ عينة تربة مسمدة مجمعة من أعماق صفر-٢٥ سم على طبقات بسمك ٥ سم. حيث تراوح النشاط الأشعاعى فى العينات التى تم دراستها وفحصها بين ٦,٠± ١,٢و ٨٧,٥ ± ١,٢ بمتوسط ٣١,١٢ ± ٢,٢٢ بيكريل/كجم للراديوم-٢٢٦، وبين ٣,٨ ± ١,٢ و ١٤,٢ ± ٣٣ بمتوسط ١٠,٩٦ ± ١,٨٩ بيكريل/كجم للثوريوم-٢٣٢، وبين ٧١,٨ ± ٢٤,٠ و ٥٤٣,٢ ± ٢٦,٥ بمتوسط ٢٦٤,١٠ ± ١١,٩٤ بيكريل/كجم وذلك للبوتاسيوم-٤٠.

كما تم أيضاً حساب الجرعة الممتصة فى الهواء فى المزرعة المسمدة والتى وجد ان قيمتها ٣١ نانوجراى/ساعة، وهى قيمة اقل من المتوسط العالمى المقدر ب٥٧ نانوجراى/ساعة. وكذلك تم تعيين الراديوم المكافئ (Ra_{eq}) ومعامل الخطورة الخارجى (H_{ex}) والجرعة السنوية المكافئة، وقورنت بالقيم العالمية الموصى بها دولياً وهى فى حدود المسموح به.