Natural Radioactivity and Its Environmental Implications in Soil Samples Collected Around Abou-Zabal Phosphate Fertilizer Factory Area in Egypt

S.Fares, W.M.Moslem, A.K.Hassan, A.A.Eltawil, F.Abdelhamied

Abstract- A total of twenty three composite soil samples was collected from different geographical areas around the Abou-Zabal phosphate fertilizer factory region area in Egypt. The aim of the present work is to determine the levels of some natural radioactivity in soil samples were collected. Activity concentration of background radionuclides such as ²²⁶Ra, ²³²Th and ⁴⁰K of these samples was determined by using a high-resolution co-axial HPGe gamma-ray spectrometry. According to the results this analysis, ²²⁶Ra was found in concentrations mean of 283.87 Bq kg⁻¹; 232 Th in mean concentrations of 16.15, Bq kg⁻¹; and 40 K in mean concentrations of 146.36 Bq kg⁻¹ for the analyzed soil, respectively. Radon (222Rn) mean concentration content in the samples, were estimated using solid state nuclear track detectors (CR - 39). The Radon gas mean concentration was found 162.37 Bq.m⁻³. Based on the radioactivity levels determined, the gamma-absorbed dose rates in soil sample above the ground were calculated. To evaluate the radiological hazard of the natural radioactivity, the radium equivalent activity, the gamma-absorbed dose rate and the mean population weighted dose rate were calculated.

Index Terms: Radioactivity concentration, Phosphate Fertilizer Factory, Soil, pollution.

I. INTRODUCTION

The contaminants for Phosphate fertilizer factory of wind borne contained in and phosphate mineral particulates emitted into the atmosphere from the fertilizer industry. These effects include the extent of soil enrichment in trace elements relative to the regional background and threshold values, and the associated hazards imposed on the soil environment. Such hazards are inherent in the mobility of the contaminants, which depends on natural radioactive elements carriers as well as on their behavior and interaction with soil constituents. Hence, total concentrations of natural radioactive elements in soils surrounding the factory, and their natural radioactive element species in the

S.Fares, Department of Radiation Physics, National Center for Radiation Research and Technology NCRRT, Atomic Energy Authority, Cairo, Egy.

Department of Physics , Faculty of Science, Baha University, Saudi Arabia

A.A.Eltwil, Armed forces Main chemical laboratory Egypt

F.Abdelhamied, Department of Physics , Faculty of Science, Port Said University Egypt.

contamination source, were determined. The specific activities of the natural radionuclides in the phosphate industrial products are depending on their based concentrations in the phosphate raw materials (phosphate rocks and/or phosphoric acid) and on their radiochemical partitioning during manufacture processes, which is affected by manufacturing (wet or thermal) procedure [1]. The raw material used in production of some fertilizers is phosphate ore containing various amounts of natural radioactive elements. During phosphate ore processing, owing to chemical properties of Radium, practically all ²²⁶Ra gets incorporated into phosphogypsum and remains in disequilibrium status when compared to radioactivity levels contained in the raw material. Most of the phosphogypsum is considered waste and is stockpiled or discharged into the aquatic environment [2]. Potential issues of concern resulting from phosphogypsum disposal are its environmental impacts; possible increases in radionuclides in soils or in groundwater and consequential ingestion by humans through exposure routes such as drinking water and food chain [3]. Once deposited in bone tissue, ²²⁶Ra has a high potential for causing biological damage through continuous irradiation of human skeleton over many years and may induce bone sarcoma [4].

Numerous types of human practices and non-nuclear industries contribute to further concentrate some of the natural radionuclides that can be found in the earth's crust affecting the human and the environment. Fertilizer industries, specifically the phosphate fertilizer industries, are important sources of exposure to ionizing radiation of the people and possible contamination of the environment as phosphate fertilizers, raw materials and by-products contain elevated level of radionuclides of ²³⁸U, ²³²Th decay series as well as radioisotopes of ⁴⁰K [5]. The production process of phosphate fertilizer redistributes radionuclides throughout the environment and introduces them into the final products and byproducts. Natural radioactivity in phosphate fertilizer factors are principally due to raw materials (phosphate rock), dehydrated calcium sulphate (phosphogypsum) generated as by-product during production of phosphate fertilizer [6]. The radium and uranium tend to separate in the process of producing phosphoric acid, an important step in fertilizer manufacture. Phosphoric acid is the starting material for triple superphosphate (TSP), single superphosphate (SSP), monoammonium phosphate (MAP) and diammonium phosphate (DAP), NPK (Nitrogen-Phosphorus-Potassium) fertilizer and di-calcium phosphate (DCP). The aim of this



W.M.Moslem, Department of Physics , Faculty of Science, Port Said University Egypt

A.K.Hassan, Department of Physics , Faculty of Science, Baha University, Saudi Arabia

study is to investigate the effects on the surrounding the Abu-Zabal phosphate fertilizer factory soils of wind borne contaminants contained in and phosphate mineral particulates emitted into the atmosphere from the fertilizer industry.

Radon is a gas heavier than air and is called (radium emanation). Since radon, is a colorless, odorless, tasteless and invisible radioactive gas, therefore suitable detectors should be used to detect its presence. ²²²Ra is a natural radioactive gas comes from the decay of ²²⁶Ra in the ²³⁸U series. Radium is distributed in soil rocks, ocean waters and sediments. In the present work the solid state nuclear track detectors (CR – 39) counting technique was used to determine both ²²²Rn and ²²⁶Ra concentrations in the samples, by measuring ²²²Rn concentrations in the sample at different intervals of time after preparation [7].

II. EXPERIMENT AND METHODLOGY

A. The study areas

The phosphate fertilizer factory is located in Abou-Zabal area between latitudes 31.36903 & 31.38784 and between longitudes 30.26522 & 30.28175. The factory is around 30 km from Cairo. The factory is surrounded by farmlands, owned by farmers. Ismailia Lake is the main source of water for.



Fig (1). Site Collecting soil samples around Abou-Zabal fertilizer factory.

B. Soil sampling and preparation

A total of twenty three composite soil samples were collected around Abou-Zabal phosphate fertilizer factory in Egypt . Multiple samples were collected from 0-5 cm depth at a location and homogenized to make one representative sample. As shown in Figure 1. A total of 22 soil samples, each about 2 kg in weight, were crushed, homogenized and sieved to about 100 mesh by a crushing machine. The samples were then placed for drying at 105 °C for 24 h to ensure that moisture is completely removed. The samples were transferred to beakers polyethylene container of 250 cm3 volume. Each sample was carefully sealed for four weeks to reach secular equilibrium between 232Th and 238U and its short lived daughter products [8].

C. Detectors calibration

1- HPGe detector

The soil samples from collected around Abou-Zabal phosphate fertilizer factory were analyzed using a highresolution; low-back ground gamma-ray spectrometry system based on a coaxial high purity germanium detector (HPGe). The diameter of the crystal is 62.10 mm and was operated under a high voltage, bias of (+) 3000 V (DC). The unwanted ambient radioactivity from room background sources was reduced by surrounding the detector with a cylindrical lead shield of about 10 cm thickness. The gamma-ray spectra which were analyzed were created through converting the event energy into a pulse height spectrum. The signal processing was done by connecting the detector to a preamplifier and a standard spectroscopy shaping amplifier. The resultant spectral was analyzed using Canberra Genie software "Genie-2000" [9].

The performance of the HPGe detector used in the current work was characterized by the energy calibration and its absolute full energy peak detection efficiency. The initial energy calibration of the detector was carried out using full-energy peaks from a mixed ¹³³Ba and ¹⁵²Eu standard source with activity of 9.33 ± 0.22 kBq⁻¹ and 18.46 ± 0.48 kBq⁻¹ respectively. The spectrum was acquired for 1200s for energy calibration while same standard was measured for 21600 sec. for efficiency calibration. The range of energies included 81.00 keV to 1407.92 keV corresponding to channels 246 - 4424 respectively for the initial energy scale of the program. In order to avoid any drift in the later soil samples' measurements, the system was recalibrated on a weekly basis during the sample measurements. The absolute efficiency calibration of the detector was carried out using full energy peaks from the aforementioned mixed standard sources. The source was placed surrounding the germanium detector with the radionuclides dispersed in gel matrices within the Marinelli beaker of geometries identical to those of the evaluated samples. The efficiency calibration spectrum was acquired for 21600 sec. and a range of discrete gamma-ray energies from 81.00 keV from ¹³³Ba up to 1460.81 keV from ¹⁵²Eu were used. An empty sealed Marinelli beaker with the same geometry was used on a weekly basis during the measurement period to determine the background spectrum observed by the germanium detector. The counting time of the ambient background spectrum was also 43200 sec [10].

2- CR-39 detector

The concentration and exhalation rate of radon can be made using CR-39 detectors because of their capability to register tracks at different levels of registration sensitivity. The calibration of CR-39 should be performed where the integrating radon's concentration is known [11]. The CR-39 detectors used in this work were supplied by Pershore Mouldings, Ltd., UK, in the form of large sheets which were cut into 2 cm2 cm squares. Each sample was placed in a glass cylinder of radius 3.5 cm and 10 cm length. Dosimeters were prepared by putting two CR-39 detectors in the bottom of the chamber cover. The cylindrical container was sealed; the samples were stored for at least 30 days. The determination of the concentrations of alpha particles emitted from radon gas in soil samples were performed by using the nuclear track detector (CR-39) of thickness (250 μ m) and area of about (1×1cm2). The radon gas concentration in soil samples was obtained by using the sealed-cup technique as shown in Fig. (2). After the



irradiation time (45 day), the (CR-39) track detectors were etched in (6.25 N), (NaOH) at temperature of (60 Co) for (6 h), and the tracks density were recorded using an optical microscope with magnification (400x).The density of the tracks (ρ) in the samples were calculated according to the following relation [12].



Fig. (2): A schematic diagram of the sealed-cup technique in soil sample.

D. Samples analysis

The samples were placed directly over the front face of the detector. The acquisition time for each sample was 43200 sec. The counting geometry of the samples and the standard sources used for efficiency calibration were kept constant. A range of different gamma ray energy transition lines ranging from 74.81 keV for ²¹²Pb up to 2011 keV for ⁸ ³⁷Kr. associated with the decay products of the ²³⁸U and ²³²Th decay chains were analyzed independently to obtain more statistically significant overall results. These data were analyzed under the assumption of secular equilibrium of the radionuclides within these samples. Background contributions were subtracted from the peak areas for the measured samples. Several transitions from decays of shorter-lived radionuclides in the ²³⁸U decay chain, such as ²²⁶Ra, ²¹⁴Pb and ²¹⁴Bi (which can be thought of as ²²⁶Ra indicators), were used to estimate the weighted mean of the activity concentration of ²²⁶Ra. The weighted mean of the activity concentration of ²³²Th was determined using gamma-ray transitions associated with the decays of ²²⁸Ac, 212 Pb and 208 Tl. The activity concentration of the radionuclides found in the soil samples were determined using the following equation, and expressed in Bqkg⁻¹:

$$A = \left(\frac{C_{NP}}{g_{dI} \times \varepsilon(E_{\gamma}) \times m}\right) Bqkg^{-1}$$
⁽¹⁾

where C_{NP} = net peak counts for a given energy line, g_{dl} = absolute gamma decay intensity for the specific energy photo-peak, $\varepsilon(E_{\gamma})$ = the absolute photo-peak efficiency of the germanium detector at this energy and m is the mass of the sample in kg. The weighted mean of the activity concentration of ²²⁶Ra and ²³²Th following the decay of the shorter-lived radionuclides in ²³⁸U and ²³²Th decay chains and the associated errors were evaluated based on the following equations [12];

$$W_{mean} = \frac{\sum_{i=1}^{n} \frac{a_i}{(b_i)^2}}{\sum_{i=1}^{n} \frac{1}{(b_i)^2}}$$
(2)

Where: $a_i \mp b_i$ = respective associated activities (a_i) and b_i

uncertainty $({}^{b_i})$ of the shorter-lived radionuclides in 238 U and 232 Th decay chains, $i = 1, 2, \dots, n$

And the propagated uncertainty in the weighted mean is:

$$\Delta W_{mean} = \left(\sqrt{\sum_{i=1}^{n} \frac{1}{(b_i)^2}}\right)$$
(3)

The gamma-ray peaks associated with decays from 40K at 1460.21 keV was used to determine the activity concentrations for the nuclei. The activity concentration of ²²⁶Ra present in the samples was estimated indirectly from one of the reference peaks in radium indicators. The contribution of ²²⁶Ra through its (3.59 % branching ratio) gamma ray at the energy of 186.1 keV was calculated by employing the spectral interference correction method [13] using the least spectrally disturbed peak (reference peak) at 295.2 keV from the decay of ²¹⁴Pb. The net peak area of ²²⁶Ra at the line186.1 keV was then estimated by employing the equation [14]:

$$C_{NP at 186.1keV} (^{226}Ra) = \frac{(g_{dl})_{at 186.1keV}}{(g_{dl})_{at 295.2keV}} \left[F_{C}F_{S} \right]_{at 186.1keV}}{(F_{C}F_{S})_{at 295.2keV}} \times C_{NP at 295.2keV} (^{214}Pb)_{Ref}}$$
(4)
where
$$C_{NP at 186.1keV} (^{226}Ra)$$
are the net peak counts at 186.1
keV line for
$$^{226}Ra, C_{NP at 295.2keV} (^{214}Pb)$$
are the net peak

counts at 295.2 keV line for ²¹⁴Pb (reference peak), g_{dl} are the respective gamma decay intensity for 186.1 keV and 295.2 keV lines, ε are the respective efficiency values for 186.1 keV and 295.2 keV lines and F_C and F_S are the correction factors of the true coincidence summing peak and self- absorption. The effect of the true coincidence summing peak and self-absorption were considered to be negligible (=1) in the current work.

E. Calculation of radiological risk

The measured activity of ²³⁸U, ²³²Th and ⁴⁰K were converted into doses by applying the factors 0.461, 0.604 and 0.0417 for uranium, thorium and potassium respectively as:

$$D_{R} = (0.461 A_{R} + 0.604 A_{Th} + 0.0417 A_{k}) nGyh^{-1}$$
(5)

where D_R is the gamma dose rate in the outdoor air at 1m above the ground and A_{Ra} , A_{Th} and A_K are the activity



Natural Radioactivity and Its Environmental Implications in Soil Samples Collected Around Abou-Zabal Phosphate Fertilizer Factory Area in Egypt

concentrations (Bq kg⁻¹) of radium, thorium and potassium, respectively in the samples. In the above conversion factors, it is assumed that all the decay products of ²²⁶Ra and ²³²Th are in radioactive equilibrium [15]. The published maximal admissible (permissible) dose rate is 51 nGy/h [16]. In order to estimate the annual effective dose rate in air, the conversion coefficient from the absorbed dose in air to the effective dose received by an adult has to be taken into consideration. This value is 0.7 SvGy⁻¹ for environmental exposure to gamma rays of moderate energy published in UNSCEAR [2]. The outdoor and indoor occupancy factors are 0.2 and 0.8 respectively [18]. The annual effective dose equivalent is given by:

$$AEDE_{outdoor} = (D_R \times DCF \times 0.2 \times T) mSv / yr$$
$$AEDE_{indoor} = (D_R \times DCF \times 0.8 \times T) mSv / yr$$
(6)

Where: DCF =dose conversion factor (0.7 SvGy⁻¹) and T= time (8760 hyr⁻¹). The world average annual effective dose equivalent (AEDE) from outdoor terrestrial gamma radiation is 0.046 mSv/year. The annual effective dose external is given by the equation [22],[23]:

$$AEDE_{EX} = \left(\sum AEDE_{outdoor} + AEDE_{indoor}\right) mSv / yr$$
(7)

Excess lifetime cancer risk (ELCR) was calculated by using equation (7):

$$ELCR = AEDE_{outdoor} \times E_{LD} \times C_{RF}$$
(8)

where E_{LD} = Expected lifetime duration (70 yrs.) and C_{RF} = fatal cancer risk factor (for stochastic effects, ICRP, uses a value of 0.05 for the general public) The world average annual effective dose equivalent (AEDE) from

outdoor terrestrial gamma radiation is 0.046 mSv/year [2].

Radium equivalent activity (Ra_{eq}) is used to assess the hazards associated with materials that contain 226 Ra, 232 Th and 40 K in Bqkg-1, which is, calculated on the assumption that 370 Bqkg⁻¹ of 226 Ra or 260 Bqkg⁻¹ of 232 Th or 4810 Bqkg⁻¹ of 40 K produce the same gamma dose rate. The Ra_{eq}

 Ra_{eq} of the sample in Bqkg⁻¹ can be achieved using the equation [19]-[21]:

$$Ra_{eq} = \left[\left(A_{Th} \times 1.43 \right) + \left(A_K \times 0.077 \right) + \left(A_U \right) \right] Bq Kg^{-1}$$
(9)

The radium equivalent is the most useful guideline for regulating safety standards on radiation protection for the general public and as published, the maximal admissible

(permissible)
$$Ra_{eq}$$
 is 370 Bqkg-1 [18].

In order to evaluate the external hazard index $({}^{H}{}_{ex})$, a model proposed by Beretka and Mathew [20] was used. This index actually evaluates the hazard of the natural gamma radiation [22]. However, the prime objective of this index is to limit the radiation dose to the permissible dose equivalent limit of 1mSv/yr [23],[24]. The equation used in evaluating H

 H_{ex} is given as;

$$H_{ex} = (A_U / 370) + (A_{Th} / 259) + (A_K / 4810) \le 1$$

(10) The criterion of this model considers that the external hazard due to gamma-rays corresponds to a maximum radium-equivalent activity of 370 Bq/kg for the material [20],[26].

F. Activity Indices {Gamma-index (Iy) and Alpha Index (Ia)

A number of indices dealing with the assessment of the external and internal radiations originating from building materials and gamma concentration indices have been proposed by several investigators [26],[27]. In this study, the gamma-index was calculated as proposed by the European Commission (EC, 1999) [33]:

$$I\gamma \Box = A_{Ra}/150 + A_{Th}/100 + A_{K}/1500$$
(11)

 A_{Ra} , A_{Th} and A_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq·kg⁻¹, respectively. The mean values of I_γ calculated from the measured activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K are presented in Table (2) for different all samples and all the regions from where they were collected. So far, several alpha indices have been proposed to assess the exposure level due to radon inhalation originating from soil materials. While the values of I_γ for critical value are unity I_γ =1 [28]. The alpha index was determined using the following formula:

$$I\alpha = A_{Ra}/200 (Bq kg^{-1})$$

(12)

 A_{Ra} (Bq kg⁻¹) is the activity concentration of ²²⁶Ra assumed in equilibrium with ²³⁸U. The recommended exemption and upper level of ²²⁶Ra activity concentrations in soil are 100 and 200 Bq kg⁻¹, respectively, as suggested by [29]. These considerations are reflected in the alpha index. The recommended upper limit concentration of ²²⁶Ra is 200 Bq kg⁻¹, for which I α = 1.

G. Evaluation of ^{222}Rn

The determination of the concentrations of alpha particles emitted from radon gas in soil samples were performed by using the nuclear track detector (CR-39). The density of the tracks (ρ) in the samples were calculated according to the following relation:

$$track \cdot density(\rho) = \frac{A \text{ verage } \cdot number \cdot of \cdot total \cdot pits(tracks)}{A \text{ rea } \cdot of \cdot field \cdot view}$$
(13)

The radon gas concentration in the soil samples were obtained by the comparison between track densities registered on the detectors of the samples and that of the standard soil samples. Radon concentrations (C_{Rn}) were calculated by the formula

[30-36]

$$C_{Rn} (Bqm^{-3}) = \frac{N_0 t_0 \rho}{\rho_0 t}$$
(14)

Where: No = activity concentration for a standard source (^{226}Ra) , to = exposure time for standard source, ρ o=track density for a standard source (track cm⁻²), ρ = track density



for sample (track cm⁻²), and t = exposure time of the sample. The effective radium content of the solid sample can be calculated by the formula [37],[39]:

$$C_{Ra}(Bqkg^{-1}) = (\frac{\rho}{kT_e})(\frac{hA}{M})$$
(15)

where M is the mass of sample in kg, A is the area of a cross section of the cylindrical can in m^2 and h is the distance between the detector and the top of the sample in m. ρ is the counted track density, k is the calibration factor of the CR-39 track detector, and Te denotes the effective exposure time. The exhalation rate was calculated using the following equation [30]-[33], [34], [39],[40]:

$$E_{x} = \frac{CV \lambda}{A \left(T + \frac{e^{-\lambda T} - 1\right)}{\lambda}}$$
(16)

Where: Ex is the radon exhalation rate (Bq kg⁻¹ d⁻¹), C is the measured radon concentration by the (CR-39) detector (Bq m d⁻¹), λ is the decay constant of radon (d 1), T is the exposure time (d), V is the volume of the radon chamber (m³), and A is the mass of the sample. The annual effective dose (HE) to the personal work was calculated from the following formula according [2], [31]-[33]:

$$H_E (mSv y^{-1}) = C x F x T x D$$
(17)

where C is the radon concentration in Bq m⁻³, F is the ²²²Rn indoor equilibrium factor (0.4), T is time (8760 h y⁻¹), and D for dose conversion factor (9 x 10-6 mSv y⁻¹ (Bq m⁻³)⁻¹).

III. RESULTS AND DISCUSSION

A. Activity concentration

activity The Minimum , Maximum and mean of concentration of radionuclides in the soil samples range from 87.47 to 514.00 and 283.87 Bq.kg⁻¹ for ²²⁶Ra, 8.00 to 37.00 and 16.15 Bq.kg⁻¹ for ²³²Th and 19.00 to 179.88 and 146.36 $Bq.kg^{-1}$ for ${}^{40}K$ (Table 1). These values are higher than the world average ranges for ${}^{226}Ra$, ${}^{232}Th$ and within the world average ranges for 40 K, 35(10–50) Bq.kg⁻¹ for 226 Ra, 35(7–50) Bq kg⁻¹, for 232 Th and 370(100–700) Bqkg⁻¹ for 40 K respectively [2]. Our result shows that the ²²⁶Ra, ²³²Th and ⁴⁰K activity concentration in the soil samples decrease as the sampling distance from the fertilizer factory increases. This may be due to contribution of fallout of phosphate ore dusts generated during the processing of the phosphate ore in the immediate vicinity of the fertilizer factory. The absorbed dose rates in air due to the natural occurring radionuclides (²²⁶Ra, 232 Th and 40 K) in the soil samples were calculated based on the [2] model.

B. Annual effective dose rates

The absorbed dose rate in air due to these radionuclides range from 52.08 to 261.24 nGy.h⁻¹ with mean of 147.74 n Gy.h⁻¹. This value is higher than normal value in soils (18–93 nGy.h⁻¹) with mean of 59 nGy.h⁻¹[2]. It is however about higher than the observed activity in control soil (15.55 μ Gy. h⁻¹). To estimate the annual effective dose rates, the product

of absorbed dose, conversion coefficient $(0.7 \ \mu\text{Sv Gy}^{-1})$ and outdoor occupancy factor (0.2) was obtained [2] .The annual effective dose values range from 60.00 to 320.00 μSv with a mean of 180.00 μSv . This mean value is less than the world average value in normal soil of 480 μSv [2] but higher than 19.0 μSv observed in control soil samples.

C. Radium equivalent (Raeq) and external hazard index (Hex)

The natural radioactivity in soil is usually determined from ²³⁸U, ²³²Th and ⁴⁰K contents. Since, Radium and its daughter products produce 98.5 % of the radiological effects of the Uranium series, the contribution from the ²³⁸U has been substituted with the decay product ²³⁸U. Radium equivalent activity is an index that has been introduced to represent the specific activities of ²³⁸U, ²³²Th and ⁴⁰K by a single quantity, which takes into account the radiation hazards associated with them. The Ra_{eq} is related to the external γ -dose and internal dose due to radon and its daughters.

The radium equivalent activity (Raeq) values for all soil samples under investigation (Table1) in soil sample are from 111.29 to 568.37 Bq kg⁻¹ and are larger compared to 370 Bq kg-1, acceptable for the public [41]. The maximum value of 568.37 Bq kg⁻¹ was recorded for S0 site (inside the factory) and the minimum of 111.29 Bq kg⁻¹ was recorded for S20 site (Farther from the factory site). The result of Raeq shows that sample from S0 to S7 the values were higher than the permissible limit recommended value and another sample within the permissible limit by ICRP [18]. The average external radiation hazard index (Hex) for the soil samples were from 0.30 to1.54 and mean 0.86 respectively. These samples from S8 to S22 with values were low compared to the maximum value of unity and sample from S0 to S7 were higher the maximum value of unity which corresponds to the maximum radium activity of 370 Bq kg⁻¹ for all terrestrial material.

D. I Gamma and I Alpha

The calculated values of I γ for the studied samples varied in the range between 0.77 –3.09. The mean calculated values of I γ for the studied samples values (2.16) .While the values of I γ for the studied samples are larger than unity as its value, which were higher than the critical value of unity. The mean computed I α values for the studied samples are given in Table (2) for the different soil samples and the regions where they were collected. The values of I α ranged from (0.44 to 2.08), with the mean value of 1.42.

E. Radon exhalation ²²²Rn

The present investigation based on the study of 23 samples from different kinds of soil fertilizer. Table (3) presents radon and radium concentrations for soil fertilizer samples in different sits. The highest average radon gas concentration (292.98 to 205.98 Bq m⁻³) found in sample from S0 to S7, whereas the lowest average radon gas concentrations found in samples from S8 to S22 as shown in Table (3). The samples from S0 to S7 with results show that the radon gas concentration are highest than the allowed limit (International Commission of Radiation Protection) (ICRP) agency [18], and samples from S8 to S22 are below than this limit. The Doses from inhalation rates in the collected soil samples are given in Table (3) (103.49 to 604.16 μ Svy⁻¹), with mean of 337.6 μ SVy⁻¹. Annual effective dose HE



(mSv y⁻¹) varied from 9.31 mSv y⁻¹ to 2.33 mSv y⁻¹ (mean of 5.19 mSv y⁻¹). To characterize the risk of exposure to radon in indoor air of residences as a result of emissions from soil samples countertops, indoor air concentrations estimated from the measured radon flux (49.86 to 292.98 and mean164.54 Bq m⁻³) were compared to relevant benchmarks that included exposure guidelines for radon, background levels of radon, and quantitative estimates of health risk. The U.S. Environmental Protection Agency, for example, has established an action level of 148 Bq m⁻³ (4.0 pCi L⁻¹) for radon in residential indoor air (EPA 1993b) [42]. Similarly, the ICRP recommends an action level for radon in indoor air of dwellings that is no lower than 200 Bq m⁻³ (5.4 pCi L⁻¹) (ICRP 2005) [43].

IV. CONCLUSION

The contents of natural radionuclides (²³⁸U, ²³²Th, and ⁴⁰K) were measured in four soil samples using the high-resolution gamma-spectrometry. The data obtained for ²³⁸U and ²³²Th contents in (ppm) were compared with the results obtained by a method based on the measurements of Radon using Solid State Nuclear Track Detectors (CR-39) in the same samples. The data obtained in this work improved the suitability of the spectrometry technique for such complex samples. It is important to determine the background radiation level in order to evaluate the health hazards. This study determined the average soil activity concentrations of the samples was collected from different geographical areas around the Abou-Zabal phosphate fertilizer factory region area in Egypt and compared the results with the world average values. The activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in different soil samples as well as radiological doses and risks were established. The activity concentrations for which the members of the public could be exposed were quantified through dose calculations. Annual effective gamma doses and the lifetime risks of cancer were higher than the world's average across the entire soil samples collected from samples S0 to S7. Moreover compared to the World's average, the lifetime risk of cancer in these samples were higher for most of the location while the samples from S8 to S22 are generally lower values. The Radon doses from inhalation rates and Annual effective dose HE in the collected soil samples from sample S0 to S7 were higher than the maximum value of unity higher than. But the samples from S8 to S22 are lower the than maximum value of unity.

ACKNWLEGMENT

The authors wish to thank the staff of laboratory chemical warfare, radioactive materials department, the Egyptian Ministry of Defense for the helpful and cooperate contribution in achieving this work. The authors also thank workers the staff of Central lab. , National Center for Research and Technology NCRRT, Atomic Energy Authority, Cairo, Egypt.

V. HELPFUL

A. Figures and Tables

TABLE (1A) : Mean radionuclide's concentration



Sample	A _U (Bq/kg)	A _{Ra} (Bq/Kg)	A _{th} (Bq/Kg)	A _k (Bq/Kg)
S0	462.60 ± 44.46	514.00 ± 48.06	37.00 ± 0.19	19.00 ± 1.17
S1	410.85 ± 39.48	456.50 ± 42.68	25.93 ± 0.13	104.80 ± 1.04
S2	359.10 ± 34.51	399.00 ± 37.31	20.07 ± 0.10	138.80 ± 0.91
S 3	325.70 ± 31.30	407.12 ± 38.07	23.67 ± 0.12	146.70 ± 0.93
S4	332.20 ± 31.92	415.25 ± 38.83	22.67 ± 0.12	148.44 ± 0.94
S5	324.10 ± 31.15	405.12 ± 37.88	19.82 ± 0.10	162.10 ± 0.92
S6	316.00 ± 30.37	395.00 ± 36.93	16.70 ± 0.09	161.30 ± 0.90
S7	289.10 ± 27.78	361.38 ± 33.79	17.48 ± 0.09	131.31 ± 0.82
<u>S8</u>	278.59 ± 26.77	327.75 ± 30.64	15.56 ± 0.08	156.50 ± 0.74
S 9	264.30 ± 25.40	310.94 ± 29.07	9.91 ± 0.05	130.90 ± 0.71
S10	250.01 ± 24.03	294.13 ± 27.50	12.73 ± 0.06	142.00 ± 0.67
S11	254.99 ± 24.50	277.00 ± 25.90	12.30 ± 0.06	147.20 ± 0.63
S12	321.14 ± 30.86	305.85 ± 28.60	14.55 ± 0.07	146.60 ± 0.69
S13	185.80 ± 17.86	265.43 ± 24.82	17.00 ± 0.09	146.60 ± 0.60
S14	112.50 ± 10.81	225.00 ± 21.04	14.55 ± 0.07	146.59 ± 0.51
S15	179.42 ± 17.24	224.27 ± 20.97	13.63 ± 0.07	164.50 ± 0.51
S16	178.83 ± 17.19	223.54 ± 20.90	20.00 ± 0.10	160.00 ± 0.51
S17	181.93 ± 17.48	173.27 ± 16.20	9.79 ± 0.05	156.10 ± 0.39
S18	129.15 ± 12.41	123.00 ± 11.50	11.79 ± 0.06	179.88 ± 0.28
S19	69.45 ± 6.67	105.23 ± 9.84	9.37 ± 0.05	174.70 ± 0.24
S20	74.35 ± 7.14	87.47 ± 8.18	8.00 ± 0.04	160.80 ± 0.20
S21	112.07 ± 10.77	106.73 ± 9.98	9.45 ± 0.05	166.80 ± 0.24
S22	132.30 ± 12.71	126.00 ± 11.78	9.37 ± 0.05	174.70 ± 0.29
Mean	241.06 ± 23.17	$\textbf{283.87} \pm 26.53$	16.15 ± 1.45	146.36 ± 6.9
Max	462.60	514.00	37.00	179.88
Min	69.45	87 47	8.00	19.00

TABLE (1B) : MEAN RADIONUCLIDE'S CONCENTRATION

	5	AEDE		AEDE
	D	outdoor	EL CD	indoor
Raeq (Bq/Kg)	(nGy \h)	(mSv y)	ELCR	(mSv\y)
568.37	261.24	0.32	1.12	1.28
501.65	231.38	0.28	0.99	1.14
438.39	202.59	0.25	0.87	0.99
452.27	208.91	0.26	0.90	1.02
459.09	212.11	0.26	0.91	1.04
445.95	206.23	0.25	0.89	1.01
431.30	199.59	0.24	0.86	0.98
396.48	183.29	0.22	0.79	0.90
362.05	167.61	0.21	0.72	0.82
335.20	155.27	0.19	0.67	0.76
323.27	149.71	0.18	0.64	0.73
305.92	152.37	0.19	0.65	0.75
337.94	156.45	0.19	0.67	0.77
301.02	139.30	0.17	0.60	0.68
257.09	119.10	0.15	0.51	0.58
256.43	118.94	0.15	0.51	0.58
264.46	122.37	0.15	0.53	0.60
199.29	92.64	0.11	0.40	0.45
153.71	71.65	0.09	0.31	0.35
132.08	61.72	0.08	0.26	0.30
111.29	52.08	0.06	0.22	0.26
133.09	62.13	0.08	0.27	0.30
152.85	71.32	0.09	0.31	0.35
318.23	147.74	0.18	0.63	0.72
568.37	261.24	0.32	1.12	1.28
111.29	52.08	0.06	0.22	0.26

TABLE (2) : I ALPHA, I GAMMA AND HAZARD EXTERNAL, INTERNAL DOSE

Sample	Igamma	I _{alpha}	H _{ex}	H _{in}
S ₀	3.81	2.57	1.54	2.93
S_1	3.37	2.28	1.36	2.59
S_2	2.95	2.00	1.18	2.26
S_3	3.05	2.04	1.22	2.32
S_4	3.09	2.08	1.24	2.36
S ₅	3.01	2.03	1.21	2.30
S_6	2.91	1.98	1.17	2.23
S_7	2.67	1.81	1.07	2.05
S ₈	2.44	1.64	0.98	1.86
S ₉	2.26	1.55	0.91	1.75
S ₁₀	2.18	1.47	0.87	1.67
S11	2.22	1.50	0.89	1.70
Su	2.28	1 53	0.91	1 74

S ₁₃	2.04	1.33	0.81	1.53
S ₁₄	1.74	1.13	0.69	1.30
S ₁₅	1.74	1.12	0.69	1.30
S ₁₆	1.80	1.12	0.71	1.32
S ₁₇	1.36	0.87	0.54	1.01
S ₁₈	1.06	0.62	0.42	0.75
S ₁₉	0.91	0.53	0.36	0.64
S ₂₀	0.77	0.44	0.30	0.54
S ₂₁	0.92	0.53	0.36	0.65
S ₂₂	1.05	0.63	0.41	0.75
Mean	2.16	1.42	0.86	1.63
Max	3.09	2.08	1.54	2.93
Min	0.77	0.44	0.30	0.54

TABLE (3): 222RN AND 226RA CONCENTRATIONS IN SOIL FERTILIZER SAMPLES

	222R _n in Air (Bq/m ³)	²²² Rn indoor	Annual effective dose HE	Doses from inhalation gas
Sample	202.00	(mSvy-1)	(mSv y ⁻)	(µsvy *)
SRow	292.98	295.15	9.31	608.16
S1	260.21	262.38	8.27	540.13
S_2	227.43	229.6	7.24	472.1
S ₃	232.06	234.23	7.39	481.71
S_4	236.69	238.86	7.53	491.32
S ₅	230.92	233.09	7.35	479.34
S ₆	225.15	227.32	7.17	467.36
S ₇	205.98	208.15	6.56	427.58
S ₈	186.82	188.99	5.96	387.79
S ₉	177.24	179.41	5.66	367.9
S ₁₀	167.65	169.82	5.36	348.01
S ₁₁	170.99	173.16	5.46	354.95
S ₁₂	174.33	176.5	5.57	361.88
S ₁₃	151.29	153.46	4.84	314.05
S ₁₄	128.25	130.42	4.11	266.22
S ₁₅	127.83	130	4.10	265.36
S ₁₆	127.42	129.59	4.09	264.49
S ₁₇	98.76	100.93	3.18	205.01
S ₁₈	70.11	72.28	2.28	145.53
S ₁₉	59.98	62.15	1.96	124.51
S ₂₀	49.86	52.03	1.64	103.49
S ₂₁	60.84	63.01	1.99	126.29
S ₂₂	71.82	73.99	2.33	149.08
Mean	162.37	164.54	5.19	337.06
Max	292.98	295.15	9.31	608.16
Min	40.86	52.03	1.64	103.40

B. REFERENCES

- Hofman, J., Leicht, R., Wingender, H.J., Worner, J., 2000. Natural radionuclide concentrations in materials processed in the chemical industry and the related radiological impact. European Commission Report EUR-19264, Directorate General Environment.
- [2] United Nations Scientific Committe on the Effects of Atomic Radiation. UNSCEAR 2000 report to the general assembly, with scientific annexes. Sources and effects of ionizing radiation. United Nations, New York, 2000.
- [3] T. P. Laich, A radiological evaluation of phosphogypsum, Health Phys., 60, 691–693, 1991.
- [4] G. Marovic and J. Sencar, 226Ra and possible water contamination due to phosphate fertilizer production, J. Radioanal. Nucl. Chem., Letters., 200, 9–18, 1995.
- [5] C. H. Saueia, B. P. Mazzilli and D. I. T. Favaro, "Natural Radioactivity in Phosphate Rock, Phosphogypsum and Phosphate Fertilizers in Brazil," Journal of Radioanalytical and Nuclear Chemistry, Vol. 264, No. 2, 2005, pp.
- [6] C. H. R. Saueia and B. P. Mazzilli, "Distribution of Natural Radionuclides in the Production and Use of Phosphate Fertilizers in Brazil," Journal of Environmental Radioactivity, Vol. 89, No. 3, 2006, pp. 229-239.
- [7] Nikezic D.; Baixeras C. and Kostic D.,1996. Sensitivity Determination and Optimization of a Cylindrical Diffusion Chamber, for Radon Measurements, with a CR-39 Detector. Nuclear Instruments and Methods in Physics Research A., Vol.373, 290p.

ISSN:2454-4116, Volume-2, Issue-4, April 2016 Pages 115-122

International Journal of New Technology and Research (IJNTR)

- [8] Beretka J., Mathew J. Natural radioactivity of Australian building materials. Industrial wastes and by-products, Health Phys. 48, 87-95, 1985.
- [9] Genie[™] 2000 Spectroscopy Software Operations manual (9233652F V3.1)
- [10] Ian Rittersdorf 2007. Gamma Ray Spectroscopy. Nuclear Engineering & Radiological Sciences. March 20, 2007
- [11] Khan, A.J., Varshney, A.K., Parsed, R., Tyagi, R.K., Ramachandran T.V., 1990. Calibration of CR-39 plastic track detector for the measurement of radon and its daughters in dwellings. Nucl. Tracks Radiat. Meas. 17, 497–502.
- [12] Esparon A & Pfitzner J (2010). Visual gamma: eriss gamma analysis technical manual. Internal Report 539, December, Supervising Scientist, Darwin.
- [13] De Corte,F., Umans,H., Vandenberghe,D. ,DeWispelaere,A.,etal., (2005).Direct gamma spectrometric measurement of the 226Ra 186.2keVlinefordetecting 238U/226Ra disequilibrium in determining the environmental dose rate for the luminescence dating of sediments. Appl. Radiat. Isot. 63 (5–6),589–598.
- [14] Yucel, H., Solmaz,A.N., Kose, E., Bor,D., (2009). Spectral in terference corrections for the measurement of 238U in materials rich in thorium by a high resolution [gamma]-ray spectrometry. Appl. Radiat. Isot. 67 (11), 2049–2056.
- [15] Singh, Surinder., Rani, Asha, Mahajan., Rakesh Kumar. (2005). 226Ra, 232Th and 40K analysis in soil samples from some areas of Punjab and Himachal Pradesh, India using gamma ray spectrometry. Radiat. Meas. 39(4), 431–439.
- [16] Olise, F. S., Owoade, O. K., Olaniyi, H. B. and Obiajunwa, E. I., 2010. A Complimentary Tool in the Determination of Activity Concentrations of Naturally Occurring Radionuclides. Journal of Environmental Radioactivity 101, 910-914.
- [17] Saito, J. (2003). Natural radionuclides in volcanic activity. Applied radiation and isotopes, 58(3), 393-399.
- [18] 18- ICRP (1990). Recommendations of the International Commission on Radiological Protection. ICRP Publication 60. Annals of the ICRP. Pergamon Press, Oxford, UK
- [19] UNSCEAR, (1982). Ionising Radiation: Sources, and Biological Effect. United Nations Scientific Committee on the Effect of Atomic Radiation, United Nations, New York, ISBN: 9211422426.
- [20] Beretka, J., and Mathew, P. J. (1985). Natural radioactivity of Australian Building materials, industrial wastes and by-products, Health Phys. Vol. 48.
- [21] Kumar, Ajay, Kumar, Mukesh, Singh, Baldev, Singh, Surinder, (2003). Natural activities of 238U, 232Th and 40K in some Indian building materials. Radiat. Meas. 36(1–6), 465–469.
- [22] Al-Ghorabie, Fayez H.H., (2005). Measurements of environmental terrestrial gamma radiation dose rate in three mountainous locations in the western region of Saudi Arabia. Environ. Res. 98 (2), 160– 166.
- [23] Al-Hamarneh, Ibrahim F., Awadallah, Mohammad I., (2009). Soil radioactivity levels and radiation hazard assessment in the highlands of northern Jordan. Radiat. Meas. 44 (1), 102–110.
- [24] Amrani, D., Tahtat, M., (2001). Natural radioactivity in Algerian building materials. Appl. Radiat. Isot. 54 (4), 687–689.
- [25] El-Kassas, I.A., Geostatistical, A, (1991). Study of gamma radioactivity at some anomalous localities in Qatar Peninsula, the Arabian Gulf. J. King Saud Univ. 4 (Science (1)), 101–112.
- [26] E. Stranden, Some aspects on radioactivity of building materials. Physica Norvegica 8, 167–173. 1976.
- [27] M. Markkanen, Radiation dose assessments for materials with elevated natural radioactivity. Report STUK-B-STO 32, Radiation and Nuclear Safety Authority e STUK. 1995.
- [28] European Commission (EC), Radiation Protection 112. Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials, Directorate-General Environment, Nuclear Safety and Civil Protection, 1999.
- [29] ICRP, Protection against Rn-222 at home and at work. Publication No. 65; Ann. ICRP 23 (2), Pergamon, Oxford, 1994.
- [30] Almayahi, B. A., Tajuddin, A. A., Jaafar, M.S, 2012. 210Pb, 235U, 137Cs, 40K, and 222Rn concentrations in soil samples after 2010 Thai and Malaysian floods. Advan. Biomed. Engin. 6, 593-598.
- [31] Almayahi, B. A., Tajuddin, A. A., Jaafar, M.S. (2014). Calibration technique for a CR-39 detector for soil and water radon exhalation rate measurements. Radioanal Nucl Chem 301:133–140.
- [32] Ali K. Alsaedi, B.A. Almayahi, A.H. Alasadi, 2013. Cement 222Rn and 226Ra concentration measurements in selected samples from different companies. Asian Journal of Natural and Applied Sciences (AJSC), 2, 4, 95-100.



- [33] Almayahi B.A., Asaad K. Alsaedi, Ali K. Alsaedi, A.H. Alasadi, Azhar S. Ali, Raad. O. Hussein, Hayder. H. Hussain, 2014. Ceramic Rn222 exhalation rates from different countries, International Journal of Scientific and Engineering Research,5; 2162-2164.
- [34] Almayahi, B.A., Asaad K. Alsaedi, Ali K. Alsaedi, A.H. Alasadi, Azhar S. Ali, Raad. O. Hussein, Hayder. H. Hussain. Bricks Rn222 Exhalation Rates in Some Samples from Different Countries. World Applied Sciences Journal, 29 (6): 706-709, 2014.
- [35] Myrick, T., Berven, B., Haywood, F. (1983). Determination of concentrations of selected radio nuclides in surface soil in the U.S. Health Phys 45, 631-642.
- [36] Mayya, Y. S., Eappen, K. P., Nambi, K. S. (1998). Methodology for mixed field inhalation in monazite areas using a twin-cup dosimeter with three-track detector. Radiat Prot Dosim., 77, 177–184.
- [37] 37- Mahur, A. K., Khan, M. S., Naqvi, A. H., Prasad, R., Azam, A. (2008). Measurement of effective radium content of sand samples collected from Chhatrapur beach, Orissa, India using track etch technique. Radiat Meas., 43, S520–S522.
- [38] Khan, M. S., Zubair, M., Verma, D., Naqvi, A. H., Azam, A., Bhardwaj, M. K. (2011). The study of indoor radon in the urban dwellings using plastic track detectors. Environ Earth Sci., 63, 279– 282.
- [39] Durrani S., Ilic, R., (1997). Radon Measurements by Etched Track Detectors. Applications in Radiation Protection, Earth Sciences, and the Environment (Singapore: World Scientific).
- [40] Grasty, R. (1997). Geophysics, 62, 1379–1385.
- [41] 41- NEA-OECD, Nuclear Energy Agency. Exposure to radiation from natural radioactivity in building materials, Report by NEA Group of Experts, OECD, Paris.1979
- [42] EPA. 1993b. Radon Mitigation Standards. Washington, DC: U.S. EnvironmentalProtection Agency, Office of Air and Radiation.
- [43] ICRP. 2005. 2005 Recommendations of the International Commission on Radiological Protection: Summary of the Recommendations (Draft). Stockholm, Sweden: International Commission on Radiological Protection.

