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Natural radioactivity levels in phosphate fertilizer and its environmental implications in Assuit governorate, Upper Egypt

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The contents of natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were measured in phosphate and super phosphate used and product in Assuit fertilizer factory in upper Egypt by using low level gamma spectrometry. Phosphate is the main material used for the fertilizers. The phosphate samples were collected from El-Sebayia, Abo Tartour and El-Shaghab. The results are discussed and compared with the levels in phosphate rocks from different countries. The average values obtained for ^{226}Ra, ^{232}Th and ^{40}K activity concentration in different types of phosphate are lower than the corresponding global values reported in UNSCAIR publications. The obtained results show that the averages of radiation hazard parameters for the phosphate and super phosphate are lower than the acceptable level 370 Bq.kg^{-1} for radium equivalent Ra_{eq}, 1 for level index $I\gamma r$, the external hazard index $H_{ex} \leq 1$ and 59 (nGy.h^{-1}) for absorbed dose rate.

Keywords: Natural radioactivity, Phosphate, Fertilizer, Radiation hazards

1 Introduction

The fertilizers play a vital role in agriculture, this led to the spread of the industry of fertilizers in all over the world widely and also to the extraction of raw materials of phosphate which are appearing on the surface of the earth. It is a necessary to know the radioactivity of these materials keeping in viewof human's health. Therefore, Assuit fertilizers (raw materials and products) like super phosphates have been studied.

The world is naturally radioactive, and around 90% exposure to human radiation arises from natural sources such as cosmic radiation, exposure to radon gas, and terrestrial radiation. Exposure to many of the forms of natural radiation have been studied. The earth contains numerous radioactive elements; while others are continuously produced through nuclear reactions in the universe. Among the former elements, the most abundant are potassium-40 and the radioisotopes of the natural series of uranium, actinium and thorium including the parent nuclei ^{235}U, ^{238}U and ^{232}Th and the decay products from the successive alpha or beta decays while the most abundant of the cosmogenic[1] origin nuclei are ^{14}C, ^{10}Be, ^{26}A. Uranium and its decay products are found in phosphate rocks of sedimentary origin[2]. Now, these phosphates are largely used for the production of phosphoric acid and fertilizers. Their radioactivities in geo- and biosheres[3] result in health problems from radiation due to the industrial processes of preparation of fertilizers.

Phosphate is the main material used for the fertilizers. The radioactivity of the phosphate used should be checked which is coming from different places. The activity concentrations of radium-226, thorium-232 and potassium-40 were measured for phosphate samples, which is collected from Abo Tartour, El-Shaghab and El-Sebayia and super phosphate as final products in fertilizer industry. For the sake of comparison, the results of concentration levels and radiation equivalent activities are compared with similar studies carried out in other countries.

2 Experimental Details

Twenty phosphate samples were collected for investigation: 15 phosphate samples from (El-Sebayia, Abo Tartour and El-Shaghab). and 5 super phosphate samples as final product in Assuit fertilizer factory in Upper Egypt. Each sample about 1 kg in weight was washed in distilled water and dried in an oven at about 110°C to ensure that moisture is completely removed, The samples were crushed, homogenized and sieved through 200 mesh, which is the optimum size enriched in heavy minerals. Weighted samples were placed in polyethylene beaker

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of 350 cm^3 volume each. The beakers were completely sealed for 4 weeks to reach secular equilibrium where the rate of decay of the daughters becomes equal to that of the parent. This step is necessary to ensure that radon gas confined within the volume and the daughters will also remain in the sample.

2.1 Instrumentation and calibration

Activity measurements were performed by gamma ray spectrometer, employing a scintillation detector 3″×3″. It is hermetically sealed assembly which includes a NaI(Tl) crystal, coupled to PC-MCA Canberra Accuspes. To reduce gamma ray background, a cylindrical lead shield (100 mm thick) with a fixed bottom and movable cover shielded the detector. The lead shield contained an inner concentric cylinder of copper (0.3 mm thick) to absorb X-rays generated in the lead. In order to determine the background distribution in the environment around the detector, an empty sealed beaker was counted in the same manner and in the same geometry as the samples. The measurement time of activity or background was 43200 s. The background spectra were used to correct the net peak area of gamma rays of measured isotopes. A dedicated software program from Canberra has carried out the online analysis of each measured γ-ray spectrum[4].

The efficiency calibration curve was made using different energy peaks covering the range up to ≈2000 keV. Measurements were performed with calibrated source samples, which contain a known activity of one or more gamma-ray emitters of the radionuclides ^{60}Co (1173.2 and 1332.5 keV), ^{133}Ba (356.1keV), ^{137}Cs (661.9 keV) and ^{226}Ra (1764.49 keV). With certified accuracies of <2% supplied by PTB Braunschweig, Germany. Eq. (1) is used for calculating the absolute efficiency:

$$E_{\mathrm{ff}} = \frac{100 \cdot N_{\mathrm{p}}}{I_{\gamma} \cdot TOC \cdot A_{\mathrm{BOC}}} \qquad \ldots(1)$$

where N_{p} is the net peak area (count/S) at E_{γ}, I_{γ} the intensity of emitted γ-ray(%), *TOC* the time of counting(S), and A_{BOC} is the activity (Bq) of the standard source at beginning of counting (*BOC)*. A_{BOC} was calculated by Eq. (2)

$$A_{\mathrm{BOC}} = A_{\mathrm{DOC}} \exp\left(-\lambda \cdot (BOC - DOC)\right) \qquad \ldots(2)$$

where A_{DOC} is the activity (Bq) of the standard source at date of calibration *DOC* and $\lambda(s^{-1})$ is the decay constant.

Daily efficiency and energy calibrations for each sample measurement were carried out to maintain the quality of the measurements. The absolute efficiency of the detector was calculated at the specific energy of the standard sources for the same geometry of the samples. But, the γ-spectra of the samples have different γ-energies. So, we need some fitting function to calculate the absolute efficiency for any considered γ-energy. We used for this purpose a function[5] for calculating the absolute efficiency at any gamma-energy of interest in the energy range below 2000 keV, which is in the following form:

$$\eta = a - b \times \exp(-c \times E_{\gamma}^{\mathrm{d}}) \qquad \ldots(3)$$

where E_{γ} is energy in MeV, *A,b,c* and *d* are coefficient data. By Eq. (3), we can determine the absolute efficiency, η, at any specific energy E_{γ} if we know the energies and coefficient data. From the experimental efficiency curves, the coefficient data were determined by using the curve-fitting program[6]:

2.2 Uncertainty of efficiency

The combined standard uncertainty of absolute efficiency $u(EFF)$ consists of $u(N_{\mathrm{p}})$, $u(I_{\gamma})$, $u(TOC)$ and $u(A_{\mathrm{BOC}})$ so:

$$\left[\frac{u(EFF)}{EFF}\right]^2 = \left[\frac{u(N_{\mathrm{p}})}{N_{\mathrm{p}}}\right]^2 + \left[\frac{u(I_{\gamma})}{I_{\gamma}}\right]^2 + \left[\frac{u(TOC)}{TOC}\right]^2 + \left[\frac{u(A_{\mathrm{BOC}})}{A_{\mathrm{BOC}}}\right]^2 \qquad \ldots(4)$$

Because $u(TOC) << TOC$, we neglected $u(TOC)$. The value of $u(A_{\mathrm{BOC}})$ was calculated by Eq. (5):

$$\left[\frac{u(A_{\mathrm{BOC}})}{A_{BOC}}\right]^2 = \left[\frac{u(A_{\mathrm{DOC}})}{A_{\mathrm{DOC}}}\right]^2 + (BOC - DOC)^2 \cdot u^2(\lambda) \qquad \ldots(5)$$

We got $u(N_{\mathrm{p}})$ from the code Genie 2000, while $u(\lambda)$ and $u(I_{\gamma})$ were taken from the compilation[7]. The calibration standards used had a certified accuracy of ≤ 2%. By measuring several times, it could be verified with a total uncertainty of the full-energy-peak efficiency of 5%.

2.3 Calculation of activity

Calculations of count rates for each detected photopeak and radiological concentrations (activity per mass unit or specific activity) of detected radionuclides depend on the establishment of secular equilibrium in the samples. The ^{232}Th concentration

was determined from the average concentrations of ^{212}Pb (238.6 keV) and ^{228}Ac (911.1 keV) in the samples and that of ^{226}Ra was determined from the average concentrations of the ^{214}Pb (351.9 keV) and ^{214}Bi (609.3 and 1764.5 keV) decay products[8].

The activity concentration in Bqkg^{-1} (A) in the environmental samples was obtained as follows:

$$A = \frac{N_p}{e \times \eta \times m} \quad \ldots(6)$$

where N_p is the (cps) sample-(cps) B.G, e the abundance of the γ-line in a radionuclide, η the measured efficiency for each gamma-line observed for the same number of channels either for the sample or the calibration source and m is the mass of the sample in kilograms.

The uncertainty of activity $u(A)$ was calculated by the following equation:

$$u(A) = A\sqrt{\left[\frac{u(N_p)}{N_p}\right]^2 + \left[\frac{u(\eta)}{\eta}\right]^2 + \left[\frac{u(m)}{m}\right]^2} \quad \ldots(7)$$

From Eq. (7), we can find that, there are many sources of uncertainties of the activity and some may result in considerable uncertainties. The following sources of uncertainties were considered.

2.4 Uncertainty of the determination of net peak areas

The uncertainty of each single net-peak area is determined by the spectrum-evaluation code. It takes into account the Poisson uncertainties of the counts in the individual channels as well as the uncertainty of the background determination. Sometimes a peak cannot be attributed unambiguously to a single nuclide. If it seems that the contributions of other nuclides to a peak are very small, no correction was applied. Due to this procedure, a maximum inaccuracy of 2% is assumed due to contributions of other nuclides but it must be pointed out that in the average this uncertainty should be smaller. By repeated measurements, it could be verified that the total uncertainty of the efficiency calibration was 5%.

2.5 Estimation of dose rate

Conversion factors to transform specific activities A_K, A_{Ra} and A_{Th} of K, Ra and Th, respectively, in absorbed dose rate at 1 m above the ground (in nGy h^{-1} by Bqkg^{-1}) are calculated by Monte Carlo method and the values are:

$$D(n\text{Gy h}^{-1}) = 0.0417A_K + 0.462A_{Ra} + 0.604\,A_{Th} \quad \ldots(8)$$

In natural environmental radioactivity situations, the effective dose is calculated from the absorbed dose[9] by applying the factor 0.7 SvGy^{-1}:

$$\text{Indoor: Dose (nGy.h}^{-1}) \times 8.760 \times 0.8 \times 0.7\text{SvGy}^{-1} \quad \ldots(9)$$

$$\text{Outdoor: Dose (nGy.h}^{-1}) \times 8.760 \times 0.2 \times 0.7\text{SvGy}^{-1} \quad \ldots(10)$$

The annual effective dose rate outdoors in units of μ*Sv/y*, is calculated by the following formula[8]:

$$\text{Annual Effective Dose Rate} = D \times T \times F \quad \ldots(11)$$

where D is the calculated dose rate (in nGy h^{-1}), T the outdoor occupancy time (0.2×24 h ×365.25 d≈1753 h y^{-1}) and F is the conversion factor (0.7×10^{-6} Sv Gy^{-1}).

2.6 γ-ray radiation hazard indexes

The natural radioactivity of building materials is usually determined from ^{226}Ra, ^{232}Th and ^{40}K contents. As 98.5% of the radiological effects of the U series are produced by Ra and its daughter products, the contribution from the ^{238}U has been replaced with the decay product ^{226}Ra. Since sand beach minerals, rejected light sands and sea beach soils can be used in industries and building constructions, the γ-ray radiation hazards due to the specified radionuclides were assessed by three different indices[10]. Radium equivalent activity is an index that has been introduced to represent the specific activities of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them. This first index[11] can be calculated as:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad \ldots(12)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bqkg^{-1}, respectively. The Ra_{eq} is related to the external γ-dose and internal dose due to radon and its daughters. The maximum value of Ra_{eq} in building materials must be less than 370 Bqkg^{-1} for safe use. Beretka and Mathew[11] derived two other indices that represent the external and internal radiation hazards. The external hazard index is obtained from Ra_{eq} expression through the supposition that its maximum value allowed (equal to unity) corresponds to the upper limit of Ra_{eq} (370 Bqkg^{-1}). This index value must be less than unity in order to keep the radiation hazard insignificant; i.e. the radiation exposure due to the radioactivity from construction materials is limited to 1.0 mSvy^{-1}. Then, the external hazard index can be defined as:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad \ldots(13)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bqkg^{-1}, respectively.

3 Results and Discussion

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1} for phosphate and super phosphate used and produced in Assuit fertilizer factory in Upper Egypt are presented in Table 1.

From the obtained results, the following observations can be recorded:

(1) The results indicate that ^{226}Ra exhibits the highest values of activity concentration compared with ^{232}Th which may be due to the relatively higher mobility of radium with respect to thorium producing an excess in these materials of ^{226}Ra coming from other areas.
(2) The lowest values of ^{226}Ra activity concentrations are found in Abo Tartour samples while the highest values are found in El-shaghab samples.
(3) The lowest values of ^{232}Th activity concentrations are found in Abo Tartour samples while the highest values are found in El-Sebayia samples.
(4) For ^{40}K, the lower values are found in Abo Tartour samples and the higher values in El-Sebayia samples.

Table 1 — Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1} in phosphate and super phosphate

Sample number	Sample location	Ra-226	Th-232	K-40
1		242.17±14.31	78.51±3.93	171.71±8.58
2		218.35±13.61	77.06±3.86	114.65±5.73
3	Abo Tartour	282.61±15.49	78.4±3.92	119.47±5.97
4		285.99±15.7	136.46±6.83	133.37±6.66
5		267.32±14.26	84.02±4.2	106.37±5.31
Average		259.29±14.67	90.89±4.55	129.12±6.45
1		778.1±39.92	337.95±16.89	390.27±19.51
2		928.7±47.75	273.74±13.68	320.21±16.01
3	El-Shaghab	855.98±44.03	417.02±20.86	400.39±20.02
4		846.13±43.36	406±20.3	390.1±19.5
5		832.57±42.8	318.54±15.92	381.15±19.05
Average		848.3±43.57	350.65±17.53	376.42±18.82
1		1084.871±58.87	480.24±24.03	552.02±27.6
2		747.03±38.64	335.67±16.79	355.67±17.78
3	El-Sebayia	734.45±37.95	381.97±19.11	369.02±18.45
4		789.43±40.66	397.4±19.88	376.22±18.81
5		841.65±44.17	377.55±18.89	336.62±16.83
Average		839.49±44.06	394.57±19.74	397.91±19.89
1		387.76±20.91	141.98±7.09	199.91±9.99
2		465.63±25.03	120.49±6.02	182.71±9.13
3	Supper Phosphate	394.51±23.32	106.74±5.33	186.07±9.30
4		487.42±26.35	130.26±6.51	201.70±10.08
5		492.21±26.31	131.65±6.58	190.72±9.53
Average		445.50±24.38	126.23±6.31	192.22±9.61

For super phosphate, it can be seen that the activity concentration of radium-226 changes from 387.76±20.91 to 492.21±26.31 Bq.kg^{-1} with average value 445.50±24.38 Bq.kg^{-1}. Thorium-232 values are found in the range between 106.74±5.33 and 141.98±7.09 Bq.kg^{-1} with average value 126.23±6.31 Bq.kg^{-1}. While the potassium-40 activity concentrations are found in the range between 182.71±9.13 and 201.70±10.08 Bq.kg^{-1} with average value 192.22±9.61 Bq.kg^{-1}. Fig. 1 shows comparison between the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for phosphate and super phosphate. Table 2 presents comparison between the activity concentrations for ^{226}Ra, ^{232}Th, and ^{40}K in phosphate as compared with other studies.

From Fig. (1), the following observations can be recorded:

(1) The highest value of ^{226}Ra, ^{232}Th and ^{40}K activity concentration is found in phosphate and the lowest value in super phosphate.
(2) The production of fertilizers reduces the radioactivity.

So fertilizers products do not pose a significant radiological hazards when used for agriculture.

The radioactivity in phosphate varies from one country to another. The results may be important from

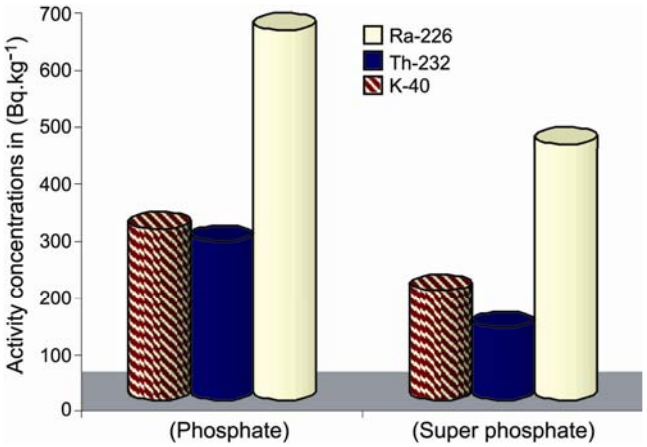

Fig. 1 — Comparison between the average values of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for phosphate and super phosphate

Table 2 — Comparison between the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $Bq.kg^{-1}$ for our phosphate with that of other countries.

Country	^{226}Ra	^{232}Th	^{40}K	References
Egypt (Abo Tartour)	259	90	129	Present work
Egypt (El-Shaghab)	848	350	376	Present work
Egypt (El-Sibaiya)	839	394	397	Present work
Egypt (El-Sibaiya)	538	25	N.F	(12)
Egypt (El-Quseir)	358	38	N.F	(12)
Egypt (Abu-Zaabal)	514	37	19	(13)
Finland	10	10	110	(14)
Pakistan (Hazara)	440	50	207	(14)
Sudan (Uro)	4131	7.5	62	(15)
Sudan (Kurun)	393	6.9	141	(15)
Tanzania (Arusha)	5022	717	286	(16)
USA (Western)	1000	20	N.F	(17)
USA (Florida)	1600	20	N.F	(17)
Morocco	1600	20	10	(17)
USSR (Kola)	30	80	40	(17)
Jordan	1044	2	8	(18)
Tunisia	821	29	32	(18)
Algeria	619	64	22	(18)

the point of view of selecting suitable materials for use in fertilizers manufacture. It is important to point out that these values are not the representative values for the countries mentioned but for the regions from where the samples were collected.

3.1 Calculation of the radiological effects

The values of radium equivalent activity Ra_{eq}, representative level index $I_{\gamma r}$, the external hazard index H_{ex} and absorbed dose rate for Abo Tartour, El-Shaghab and El-Sebayia phosphate samples used in Assiut factory in addition to the super phosphate as final product were calculated and presented in Table 3.

The highest values of radiation hazard parameters (1431.58 $Bq.kg^{-1}$, 9.80, 1.60 and 642.75 $nGy.h^{-1}$ of radium equivalent Ra_{eq}, level index $I_{\gamma r}$, the external hazard index Hex and absorbed dose rate respectively) are associated with El-Sebayia samples. These high values are due to the higher radionuclides content which consequently leads to the increase in radium equivalent activity, hence, level index $I_{\gamma r}$, the external hazard index H_{ex} and absorbed dose rate. The lowest values of radiation hazard parameters (398.31 $Bq.kg^{-1}$, 2.72, 0.37 and 180.07 $nGy.h^{-1}$ for radium equivalent Ra_{eq}, level index $I_{\gamma r}$, the external hazard index H_{ex} and absorbed dose rate, respectively) are found in Abo Tartour samples.

It is evident that for super phosphate, the radium equivalent is in the range between 560.19 to 693.80 ($Bq.kg^{-1}$) with average value 639.47 ($Bq.kg^{-1}$), the values of representative level index $I_{\gamma r}$ change between 3.82 and 4.72 with mean value 4.36. while the values of the external hazard index H_{ex} and absorbed dose rate are found in the range between 0.45 to 0.54 and 254.50 to 314.87 ($nGy.h^{-1}$) with average values of 0.52 and 290.08 ($nGy.h^{-1}$), respectively. These values are below the recommended limit by the International Commission on Radiological Protection as the maximum annual dose to members of the public. The ICRP-60, 1990 recommends[19] that any exposure above the natural background radiation should be kept as low as reasonably achievable -ALARA- but below the individual dose limits, which for radiation workers averaged over 5 years is 100 mSv and for members of the general public is 1 mSv/y. These dose limits have been established on the prudent approach by assuming that there is no threshold dose below for which there would be no effect. This means that any additional dose will cause a proportional increase in the chance of a health effect[20,21].

Table 3 — Values of radiation hazard parameters for phosphate and super phosphate

Sample number	Sample location	Ra_{eq} ($Bq.kg^{-1}$)	Dose rate ($nGy.h^{-1}$)	H_{ex}	$I_{\gamma r}$
1	Abo Tartour	366.47	166.47	0.33	2.51
2		336.58	152.2	0.32	2.3
3		403.09	182.9	0.32	2.74
4		490.47	220.11	0.55	3.36
5		394.92	178.69	0.34	2.69
Average		398.31	180.07	0.37	2.72
1	El-Shaghab	1288.69	579.88	1.38	8.82
2		1342.56	607.75	1.12	9.14
3		1480.36	664.04	1.69	10.14
4		1454.02	652.4	1.64	9.96
5		1314.76	592.94	1.3	8.99
Average		1376.08	619.4	1.43	9.41
1	El-Sebayia	1810.26	814.29	1.96	12.4
2		1251.95	562.71	1.37	8.57
3		1306.51	585.41	1.55	8.96
4		1384.05	620.43	1.61	9.48
5		1405.12	630.92	1.52	9.61
Average		1431.58	642.75	1.6	9.8
1	Super Phosphate	604.79	273.24	0.58	4.13
2		650.73	295.52	0.50	4.43
3		560.19	254.50	0.45	3.82
4		687.82	312.28	0.54	4.68
5		693.83	314.87	0.54	4.72
Average		639.47	290.08	0.52	4.36

4 Conclusions

The activity concentrations of radium-226, thorium-232 and potassium-40 were measured for phosphate and super phosphate used and final product

in Assuit fertilizer factory. These values are below the recommended limit by the International Commission on Radiological Protection as the maximum annual dose to members of the public. The radioactivity in phosphate varies from one country to another. The results may be important from the point of view of selecting suitable materials for use in fertilizers manufacture. It is important to point out that these values are not the representative values for the countries mentioned but for the regions from where the samples were collected

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