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Radon exhalation rate and Radionuclides in soil, phosphate, and building materials

S. Harb, N. K. Ahmed, Sahar Elnobi*
ERML lab,Physics Department, Faculty of Science 83523 *Qena, South Valley University, Egypt*

Abstract: *In the present study, The natural radioactivity in soil, phosphate, and building materials (sand, granite, marble, and limestone) were determined by using gamma ray spectrometer NaI (Tl) and MCA 1024. AlphaGUARD was used for radon exhalation rate. The data analyses were performed to determine ^{226}Ra, ^{232}Th and ^{40}K activity concentrations as well as ^{222}Rn exhalation rate. The radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), absorbed dose rate and annual effective dose were varied from 71.66 to 9048.94 $Bqkg^{-1}$, 0.19 to 24.45, 34.48 to 4172.34 $nGyh^{-1}$ and 0.04 to 5.12 $mSvy^{-1}$ respectively in all samples. The mass and area exhalation rates were increased from 1.31±0.09 $Bqkg^{-1}h^{-1}$ and 23.09±0.33 $Bqm^{-2}h^{-1}$, at 30 ^{0}C, to 7.98±0.56 $Bqkg^{-1}h^{-1}$ and 141.11±1.52 $Bqm^{-2}h^{-1}$, at 60 ^{0}C, respectively, for phosphate samples (grain size<80μm).*

Keywords: *Gamma-spectrometer, Alpha GUARD, Natural radioactivity, Hazard index, Radon, and Exhalation.*

I. Introduction

The natural radionuclides in soil, phosphate and building materials (sand, granite, marble, and limestone) consist mainly of ^{238}U and ^{232}Th isotopes with their daughter products as well as ^{40}K. The knowledge of the concentrations and distributions of the radionuclides in these materials enables one to assess any possible radiological risks to human health (1, 2), and provide useful information in the monitoring of environmental contamination by natural radioactivity. Nationwide surveys have been carried out to determine radium equivalent activity of building materials in many countries (3-5). The reason for current interest is due to the fact that external radiation exposures from naturally occurring radionuclides contribute, on average, of about 10% of the average annual dose to the human body from all radiation sources. It has been observed that naturally occurring radionuclides are present in soil (6-8), phosphate (9-13), building materials (14-19) which constitute a lived-in radioactive environment. In this paper, we used γ - ray spectrometry NaI(Tl) for determining the activity levels of ^{226}Ra, ^{232}Th and ^{40}K in soil, phosphate, as well as some important building materials (sand, granite, marble, limestone) commonly used in Egypt. For evaluating radon exhalation rate in the studied samples AlphaGUARD was applied. It is well known that radon exhalation of granite is greater than sand, marble, and limestone due to presence of relatively high uranium content in its natural formation (20). Radon exhalation from building materials varies with their type and origin. The knowledge of radon exhalation rate from building materials may enable the estimation of indoor radon levels. Such measurements have been the subject of many others (21-27). On the other hand, the dependency of radon exhalation rate of phosphate samples with temperatures has been studied experimentally.

II. Materials and Methods

2.1. Sampling and samples preparation

Six different types of samples have been collected from different sites in Qena and Aswan governorates, Egypt Fig (1), in order to investigate the level of activity concentration of ^{226}Ra,^{232}Th (and their decay progeny) and the primordial radionuclide ^{40}K. The samples, were air dried at room temperature in open air to ensure that any residual moisture was removed from the samples. The samples were put in an oven for 48 h at 105 ^{0}C. The soil, granite, marble and limestone samples were initially broken into coarse parts using a manual hammer. These parts were mixed using electrical sieves (FRITSCH-Germany) to obtain a homogenized material of particle size 1mm. Each samples were placed in a plastic container (250 ml), which was sealed to avoid the escape of Rn-222 and Rn-220 from the samples and left for four weeks to achieve equilibrium between^{226}Ra, ^{232}Th and their daughter products before radiometric analysis (9).

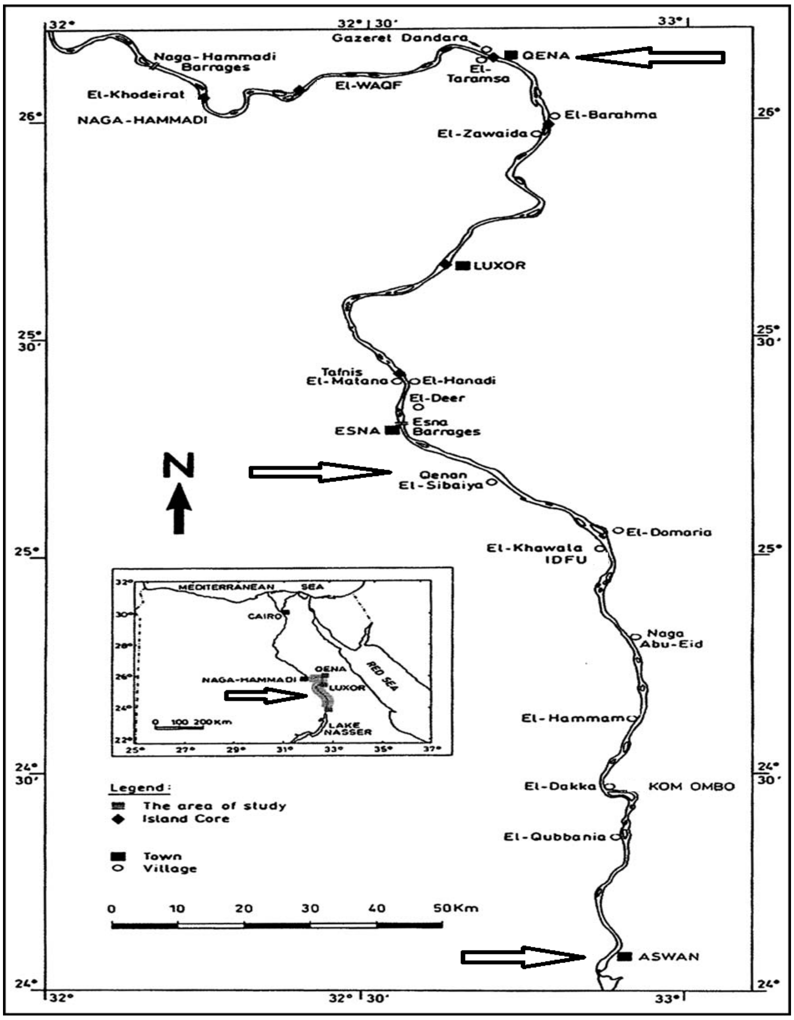

Figure 1: Sampling location.

2.2. Measuring Systems

2.2.1. Gamma-ray measurement

γ-ray measurements were carried out using NaI (Tl) detector. It consists basically of 3×3 inch NaI (Tl), S-1212-I model, with a 1024 microcomputer multichannel analyzer, 5510 Ortec Norland. The applied detector has a peak gamma ray efficiency of 2.3×10^{-2} at 1332 keV, energy resolution of 7.5% at 662 keV and operation bias voltage 805 V dc. The detector was housed inside a massive cylindrical lead shield with quarter 25 cm to reduce the background radiation. The detector is connected to preamplifier, main amplifier, analogue to digital converted (ADC) and multichannel analyzer. The system was calibrated for energy using standard point sources (^{60}Co, ^{137}Cs), and calibrated for efficiency using standard QCYB41. Every sample was placed in face to face geometry the detector for 10 to 24 hour for (^{226}Ra, ^{232}Th and ^{40}K) concentrations measurements. The resultant spectrum of each sample was acquired via the Genie 2000 software package. Prior to sampling counting, background were taken normally every week under the same condition of sample measurement. Activity concentration of ^{40}K can be measured directly by its own γ-ray at 1460.8 keV, while activities of ^{226}Ra was measured using gamma-lines at 351.92 keV (35.1%) of ^{214}Pb and at 609.32 keV (44.6%) of ^{214}Bi. ^{232}Th was determined using gamma-energies at 911.16 keV (26.6%) of ^{228}Ac and at 2614 keV (35.8 %) of ^{208}Tl. The activity levels (A_s) for radionuclides in the measured samples are computed using the following equation (28, 29):

$$A_s = \frac{\frac{N_s}{t_s} - \frac{N_B}{t_B}}{I_\gamma \cdot \varepsilon \cdot m} \tag{1}$$

Where N_s is the net number of counts in a given peak area for sample, t_s is the counting live times for sample. N_B is the net number of counts in a given peak area for background, t_B is the counting live times for background (24 hours), ε is the detection efficiency, I_γ is the number of gammas per disintegration and m is the mass in kg of the measured sample.

2.2.2. AlphaGUARD

Ionization chamber AlphaGUARD PQ2000PRO along with the additional special equipment AquaKIT was used for determining ^{222}Rn exhalation activity concentration in phosphate, soil, and building materials samples. The background of empty set-up was measured for a few minutes before every sample measuring. About 200 g of sample was put into the degassing vessel. The Alpha pump was switched on with the flow rate 1/min and 10 min flow, so ^{222}Rn activity concentration will be recorded every 10 minute. At equilibrium state, the final activity of exhaled radon inside that container is as follows:

$$A_t = A_0(1 - e^{-\lambda t}) \tag{2}$$

where λ is the decay constant of the radon nuclide and A_0 is the final value of the activity concentration, at $t \approx 7T_{1/2}$. The radon exhalation rate per unit area of the sample E_a is calculated using the following formula (30, 31).

$$Ea = A_0\lambda\ (V/F) \tag{3}$$

where V is the volume of the emanation container (2400×10^{-6} m^3) and F is the total surface area of the sample (0.0113 m^2), which equals the cross-sectional area of the emanation container. By analogy of Eq. 3, the radon exhalation rate per unit mass of that sample E_m is also calculated using the following formula:

$$E_m = A_0\lambda V/m \tag{4}$$

where, m is the mass of the sample.

III. Results and Discussions

3.1. Natural activity concentration

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K obtained from phosphate samples are 7236.2±311 Bqkg^{-1}, 906.53±55 Bqkg^{-1}, 6706.46±577 Bqkg^{-1}, respectively, and for soil, sand, granite, marble and limestone samples vary from 44.35±2 to 246.77±11 Bqkg^{-1}, 5.17±0.31 to 509.59±51 Bqkg^{-1} and 258.59±22 to 5996.64±516Bqkg^{-1}, respectively, These values are within the range of concentrations reported by UNSCEAR (32) for the radionuclides in the different samples except phosphate sample. Phosphate ores has a high content of ^{226}Ra. A summary of measurements for the activity concentration (Bqkg^{-1}) of the natural radioactivity due to ^{226}Ra, ^{232}Th and ^{40}K of different samples is given in Table 1. The distribution of ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in all samples are given in Fig. (2).

Radium equivalent activity (Raeq)

Radium equivalent activity can be calculated from the following relation defined as (33):

$$Ra_{eq} = (A_{Th}\times 1.43) + A_{Ra} + (A_K \times 0.077) \tag{5}$$

Where, A_{Th} is the specific activity of ^{232}Th in Bqkg^{-1}, A_{Ra} is the specific activity of ^{226}Ra in Bqkg^{-1}, A_K is the specific activity of ^{40}K in Bqkg^{-1}. Table 1 shows the Ra-equivalent activities calculated from Eq.5 for all samples under study.

Gamma activity concentration index (I)

A radiation hazard index, used to estimate the level of γ-radiation hazard associated with the natural radionuclides in the materials, representative level index, I, defined as (34,35):

$$I = (A_{Ra}/150) + (A_{Th}/100) + (A_K/1500) \tag{6}$$

Where, A_{Th} is the specific activity of ^{232}Th in Bqkg^{-1}, A_{Ra} is the specific activity of ^{226}Ra in Bqkg^{-1}, A_K is the specific activity of ^{40}K in Bqkg^{-1}. the result of gamma activity concentration index is shown in table 1.

Internal hazard index (H_{in})

The internal exposure to radon and its daughter products is quantified by the internal hazard index, H_{in}, which is defined as follow (33):

$$H_{in} = (A_{Ra} / 185) + (A_{Th} / 259) + (A_K / 4810) \tag{7}$$

Where, A_{Th} is the specific activity of ^{232}Th in Bqkg^{-1}, A_{Ra} is the specific activity of ^{226}Ra in Bqkg^{-1}, A_K is the specific activity of ^{40}K in Bqkg^{-1}. The result of internal hazard index is in table1

External hazard index (H_{ex})

The external hazard index, H_{ex}, is defined by some workers as:

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_K/4810) \tag{8}$$

Where, A_{Th} is the specific activity of ^{232}Th in Bqkg^{-1}, A_{Ra} is the specific activity of ^{226}Ra in Bqkg^{-1}, A_K is the specific activity of ^{40}K in Bqkg^{-1}. For safey the building materials and phosphate inside the houses and factories with respect to the workers in the phosphate factories acceptable value of H_{ex} should be less than unity (33). the result of external hazard index is given in table1.

Absorbed dose rate (D)

The gamma dose rate (D) in the outdoor air at 1 m above the ground level is calculated using the following equation:

$$D\ (nGy/h) = 0.462\ A_{Ra} + 0.604\ A_{Th} + 0.0417\ A_K \qquad (9)$$

Where, D is the dose rate in nGy h^{-1} A_{Ra}, A_{Th}, A_K are the specific activity of ^{226}Ra, ^{232}Th and ^{40}K in $Bqkg^{-1}$. The dose rates ranged from 43.87 to 4172.34 nGy h^{-1}. as could be seen in Table 1. According to UNSCEAR (2000) (32), the dose rate in air outdoors from terrestrial gamma rays in normal circumstances is ≈57 nGy h^{-1} and the national average ranges from 20 to 1100 $nGyh^{-1}$. The results obtained in this study are higher than the worldwide average of 57 $nGyh^{-1}$ due to the high ^{226}Ra concentration.

Annual effective dose (AED)

These dose rates were used to calculate an annual effective dose for each sample. As shown in Table 1, The annual effective dose equivalent can be found using the following equation

$$AED\ (\mu Svy^{-1}) = D\ (nGy\ h^{-1}) \times 8760\ h \times 0.7\ Sv.\ Gy^{-1} \times 10^{-3} \qquad (10)$$

Where, D is the dose rate in $nGyh^{-1}$, The annual effective dose are ranged from 0.05 to 5.12 $mSvy^{-1}$.

The results of the radium equivalent activity, and the calculated dose rate in air at 1 m above the ground of the present work and other studies are presented in Table 2. The average dose rate estimated for phosphate in this study is higher than the values calculated for the phosphate determined by Ref. (8, 12). The estimated dose rate for soil and building materials are comparable with the results in Bangladesh (5), and Egypt (Qena) (15). The ^{226}Ra, ^{232}Th and ^{40}K and Ra_{eq} activities of samples are higher than in other countries except soil, and limestone.

3.1. Radon exhalation

Table 3 list the radium content in $Bqkg^{-1}$, mass exhalation rate and area exhalation rate in different samples, The radium content ranged from 0.252±0.07 to 914.15±65 $Bqkg^{-1}$, and from 0.002±0.001 to 6.91±0.49 $Bqkg^{-1}h^{-1}$ for mass exhalation rate, and from 0.034±0.01 to 122.28±1.33 $Bqm^{-2}h^{-1}$ for area exhalation rate. The mean values were 223.24±16 $Bqkg^{-1}$ for radium content, 1.69±0.12 $Bqkg^{-1}h^{-1}$ for mass exhalation rate and 29.86±0.35 $Bqm^{-2}h^{-1}$ for area exhalation rate.

The maximum rate of radon exhalation rate was observed in the phosphate sample, which is 122.28±1.33 $Bqm^{-2}h^{-1}$. The lower values of area exhalation rate for radon-222 is found in marble samples, which is 0.034±0.01 $Bqm^{-2}h^{-1}$as showing in Fig.3. Fig.4 shows the correlation of the radon concentration, mass exhalation rate and area exhalation rate versus radium content of the samples under study. The linear correlation coefficient between radon exhalation rate and radium content is 1, and also between radon concentration and radium content is 0.998.

A lot of data have been published regarding to radon exhalation rates in open literature. Large discrepancy has been seen in the reported values of radium content and radon exhalation rates as can be seen in Table 4. High values are reported in Egypt by A.F. Saad (2008), M. Zubair et al. (2012), and A. Zakariya Hussein et al (2013) for phosphate, soil and sand sample, respectively whereas lower values are reported in Greece by S. Stoulos et al.(2003), Amrani and Cherouati (1999) and S. Righi (2006) for granite, limestone, and marble, respectively. Radon exhalation rates observed in the present study are well below the world average of 57. 6 Bqm^{-2} h^{-1} for all samples, except phosphate samples, and hence do not pose any health hazards to the residents.

3.3. Effect of temperature on the radon exhalation rate of the phosphate sample

The calculated values of exhalation rate for phosphate samples of the different temperature from 30 to 60^0C for a give grain size are presented in table 5.

A rise of temperature has been thought to linearly increase of radon exhalation (36). Figure 5 shows the radon exhalation rates versus temperature. A trend of increasing of the exhalation rate with temperature can be seen. There are several possible physical explanations for the presence of the temperature effect, based on the kinetic theory and thermal expansion (37). Shery (37) concluded that for typical diurnal temperature variation this effect is small. A thermally induced convection has been proposed as a factor affecting diurnal variation in exhalation (38) Shery (1983) has showed that also such an effect would not occur or at least would not be detected. and may be due to a reduction in physical adsorption of radon onto grains that occurs during the diffusion through the porous material (39).

Table 1: Average radionuclide concentrations, absorbed dose rate, effective dose, Radium equivalent activity, enteral hazard indexes, external hazard indexes, and representative level index in different samples from different location in Aswan and Qena governorates.

Type of sample	Activity concentration ($Bqkg^{-1}$)			Ra_{eq} ($Bqkg^{-1}$)	H_{ex}	H_{in}	I	Absorbed dose rate ($nGyh^{-1}$)	Effect. Dose ($mSvy^{-1}$)
	^{226}Ra	^{232}Th	^{40}K						
Phosphate	7236.2±311	906.53±55	6706.46±577	9048.94	24.45	44.01	30.89	4172.34	5.12
Soil	117.48±5	153.38±9	1521.33±131	453.95	1.23	1.54	1.67	210.81	0.26
Sand	52.15±2	21.99±1	685.79±59	136.39	0.37	0.51	0.51	66.17	0.08
Granite	246.77±11	509.59±51	5996.64±516	1437.23	3.88	4.55	5.37	673.66	0.83
Marble	67.02±3	20.85±1	222.26±19	113.96	0.31	0.49	0.40	57.65	0.07
Limestone	44.35±2	5.17±0.31	258.59±22	71.66	0.19	0.31	0.26	43.87	0.05
Min.	44.35±2	5.17±0.31	258.59±22	71.66	0.19	0.31	0.26	43.87	0.05
Max.	7236.2±311	906.53±55	6706.46±577	9048.94	24.45	44.01	30.89	4172.34	5.12
Ave.	1294±56	270±25	2565±221	1877.02	5.07	8.57	6.52	870.75	1.07

Table 2: Activity concentration in $Bqkg^{-1}$ of ^{226}Ra, ^{232}Th, ^{40}K, Radium equivalent activity ($Bqkg^{-1}$)and annual effective dose($mSvy^{-1}$)in soil, phosphate, sand, granite, marble, and limestone from different countries beside our work.

Type of sample	Activity concentration ($Bqkg^{-1}$)			Ra_{eq} ($Bqkg^{-1}$)	H_{ex}	H_{in}	I	Absorbed dose rate ($nGyh^{-1}$)	Effect. Dose ($mSvy^{-1}$)
	^{226}Ra	^{232}Th	^{40}K						
Phosphate	7236.2±311	906.53±55	6706.46±577	9048.94	24.45	44.01	30.89	4172.34	5.12
Soil	117.48±5	153.38±9	1521.33±131	453.95	1.23	1.54	1.67	210.81	0.26
Sand	52.15±2	21.99±1	685.79±59	136.39	0.37	0.51	0.51	66.17	0.08
Granite	246.77±11	509.59±51	5996.64±516	1437.23	3.88	4.55	5.37	673.66	0.83
Marble	67.02±3	20.85±1	222.26±19	113.96	0.31	0.49	0.40	57.65	0.07
Limestone	44.35±2	5.17±0.31	258.59±22	71.66	0.19	0.31	0.26	43.87	0.05
Min.	44.35±2	5.17±0.31	258.59±22	71.66	0.19	0.31	0.26	43.87	0.05
Max.	7236.2±311	906.53±55	6706.46±577	9048.94	24.45	44.01	30.89	4172.34	5.12
Ave.	1294±56	270±25	2565±221	1877.02	5.07	8.57	6.52	870.75	1.07

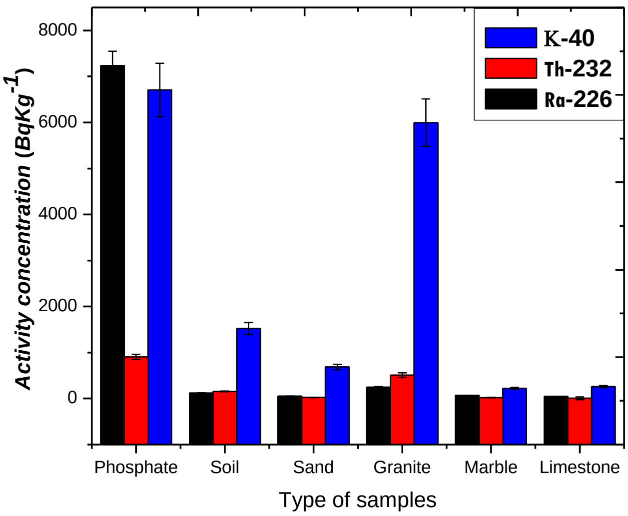

Figure 2: distribution Activity concentration (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K in different samples.

Table 3: Radon concentration, radium content and radon exhalation rate in different samples using Alpha GUARD

Type of Sample	Radon Concentration Bqm^{-3}	Radium Content $Bqkg^{-1}$	Radon Exhalation Rate	
			Mass $Bqkg^{-1}h^{-1}$	Surface Area $Bqm^{-2}h^{-1}$
Phosphate	845±29	914.15±65	6.91±0.49	122.28±1.33
Soil	261±16	279.66±20	2.11±0.15	37.41±0.47
Sand	8±3	9.25±0.8	0.07±0.006	1.24±0.05
Granite	125±11	135.46±10	1.02±0.07	18.12±0.2
Marble	0.23±0.48	0.252±0.07	0.002±0.001	0.034±0.01
Limestone	0.6±0.78	0.649±0.11	0.005±0.001	0.087±0.01
Min.	0.23±0.48	0.252±0.07	0.002±0.001	0.034±0.01
Max.	845.04±29.07	914.15±65	6.91±0.49	122.28±1.33
Ave.	206.73±10.1	223.24±16	1.69±0.12	29.86±0.35

Table 4: Radon concentration, radium content and radon exhalation rate in phosphate, soil, sand, granite, marble, and limestone from different countries beside our work

Country	Type of sample	Radon Concentration Bqm^{-3}	Radium Content $Bqkg^{-1}$	Radon Exhalation Rate		Reference
				Mass $Bqkg^{-1}h^{-1}$	Surface Area $Bqm^{-2}h^{-1}$	
Egypt (El-Sabaea)	Phosphate	845±29	914.15±65	6.91±0.49	122.28±1.33	Present Work
Egypt (El-Sabaea)		1719.6		0.658	4.125	A.F. Saad (2008)
Egypt (Qena)	Soil	261±16	279.66±20	2.11±0.15	37.41±0.47	Present Work
India			14.1	23.1	600.74	M. Zubair et al. (2012)
Syria					72–32 400	Shweikani and Hushari (2005)
Egypt (Qena)	Sand	8±3	9.25±0.8	0.07±0.006	1.24±0.05	Present Work
Malaysia		480.71 ± 4.52	9.72± 1.68	0.022	0.345	A. Zakariya Hussein et al (2013)
Egypt (Aswan)	Granite	125±11	135.46±10	1.02±0.07	18.12±0.2	Present Work
Greece				0.084 ± 0.081	1.24 ± 1.19	S. Stoulos et al.(2003)
Saudi Arabia					0.12–131	Al-Jarallah et al. (2005)
Egypt (Aswan)	Marble	0.23±0.48	0.252±0.07	0.002±0.001	0.034±0.01	Present Work
Algeri					0.035–0.066	Amrani and Cherouati (1999)
Egypt (Qena)	Limestone	0.6±0.78	0.649±0.11	0.005±0.001	0.087±0.01	Present Work
Italy					0.036 ± 0.003	S. Righi (2006)

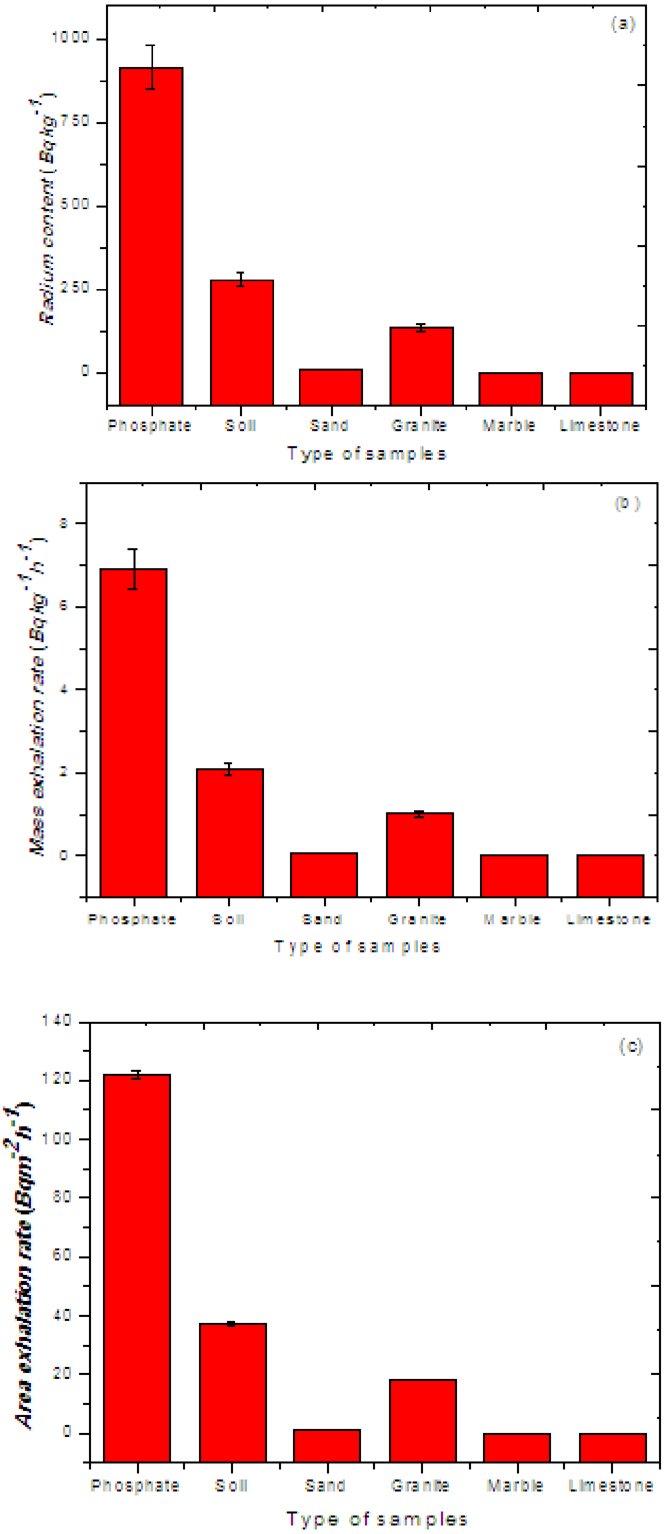

Figure 3: distribution of (a) radium content, (b) mass exhalation rate (c) area exhalation rate and type of samples.

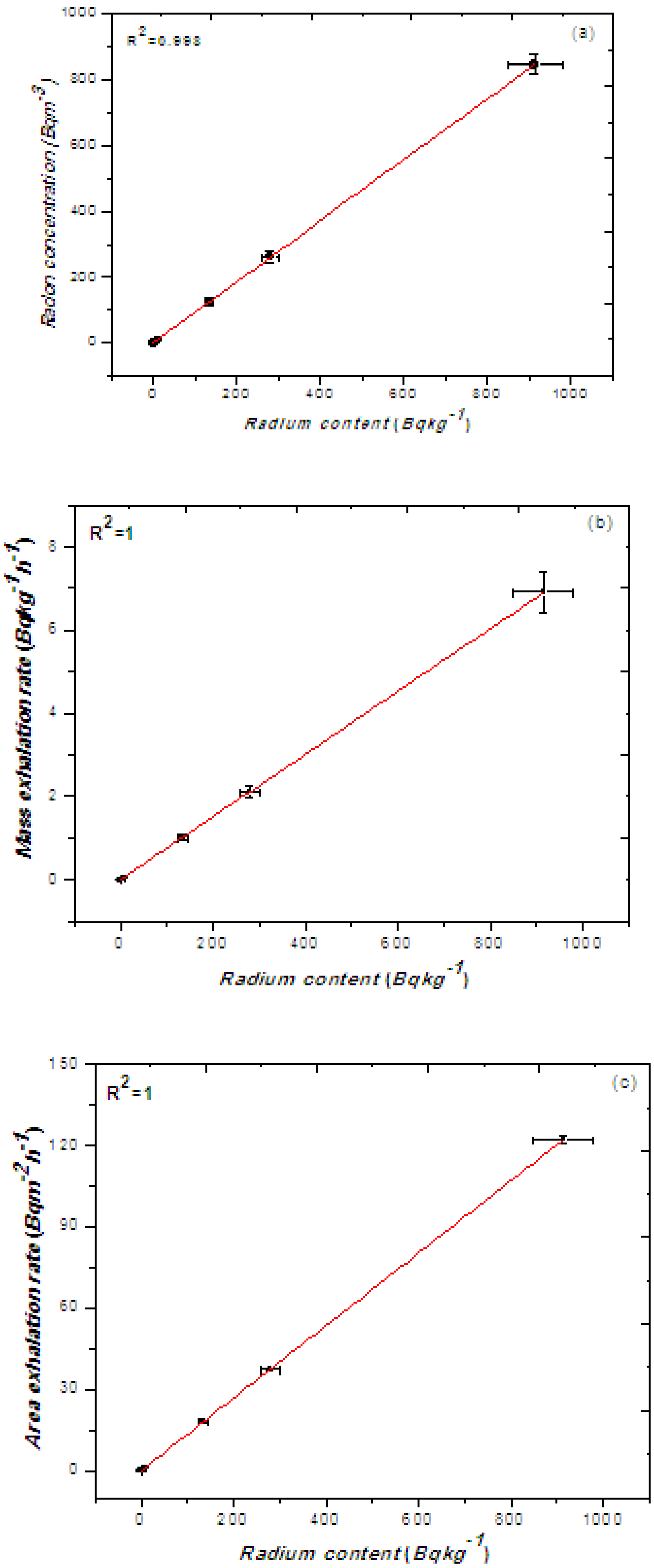

Figure 4: relation of Radium content ($Bqkg^{-1}$) & (a) radon concentration (b) mass exhalation rate($Bqkg^{-1}h^{-1}$), (c) area exhalation rate of Rn-222 ($Bqm^{-2}h^{-1}$) in different samples.

Table 5: Effect of temperature on radon exhalation rates in phosphate sample (Grain size <80μm).

Type of sample	Temperature of sample (^0C)	Radon Exhalation Rate	
		Mass ($Bqkg^{-1}h^{-1}$)	Surface Area ($Bqm^{-2}h^{-1}$)
Grain size<80□ m			
Phosphate	30	1.31±0.09	23.09±0.33
	40	1.99±0.14	35.18±0.45
	50	3.14±0.22	55.47±0.66
	60	7.98±0.56	141.11±1.52

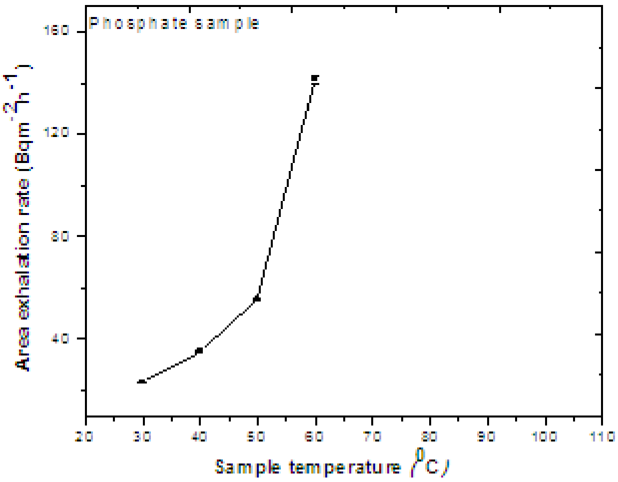

Figure 5: Area exhalation rate versus temperature of phosphate sample.

IV. Conclusion

The natural radionuclide content of soil, phosphate, and some building materials (sand, granite, marble, and limestone) commonly used in Egypt were determined. The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K are 1294±56 $Bqkg^{-1}$, 270±25 $Bqkg^{-1}$ and 2565±221 $Bqkg^{-1}$, respectively for soil, phosphate, and some building materials. The results indicate that phosphate is generally has higher natural radioactivity than other building materials and soil.

The radium equivalent activity (Ra_{eq}), internal hazard indexes (H_{in}), external hazard indexes (H_{ex}), and representative level index (I), for all samples under investigation (phosphate – soil – sand – granite – marble – limestone) were 9048.94, 453.95, 136.39, 1437.23, 113.96 and 71.66 $Bqkg^{-1}$ for Ra_{eq}, 24.45, 1.23, 0.37, 3.88, 0.31, and 0.19 for H_{in} , 44.01, 1.54, 0.51, 4.55, 0.49 and 0.31 for H_{ex}, 30.89, 1.67, 0.51, 5.37, 0.40, and 0.26 for I, respectively.

The surface exhalation and mass exhalation rates for radon from these different samples were reflected to their radium contents. The result indicate that the exhalation rates are higher for phosphate samples from El Sebaia and lower for marble samples from Qena. The overall average of mass exhalation rate for samples under study is 1.69±0.12 Bq $kg^{-1}h^{-1}$ and the average value for surface exhalation rate for radon in different samples is 29.86±0.35 Bq m^{-2} h^{-1}.

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