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Study of Hazard Indices and Radiological Doses in Phosphate Samples from El-Mahamid Area, Upper-Egypt



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IN the present study, gamma radioactivity in phosphate samples from El-Naser phosphate quarry in El-Mahamid area, Egypt was evaluated using gamma spectrometry with NaI (Tl) detector. The assessment of the radioactivity was carried out by computing several parameters of hazard indices and radiological doses for the samples analyzed in order to assess the radiological risks resulting from phosphate mining activities. The results also were compared with available similar studies and relevant reference and statistically analyzed using Statistical Program for Social Science (SPSS 25.0). The average activity concentrations of 452.06 ± 14.7 , 49.34 ± 1.68 and 360.70 ± 12.49 Bq/Kg were obtained for ^{226}Ra , ^{232}Th and ^{40}K respectively. The average activity concentration obtained for ^{226}Ra was twelve times higher than the world average value recommended by UNSCEAR. The average values of all hazard indices and radiological doses discussed in this study were found to be higher than the world average values recommended by UNSCEAR. This study provides information about radiological threat of phosphate activities on health and environment which require an additional radiological attention for protection.

Keywords: Phosphate, Spectrometry, Hazard, Radiological, UNSCEAR.

Introduction

Phosphate deposits classified into three types, phosphorites, carbonatites and alkaline igneous rocks and guano deposits. The main industrial mineral of all is apatite in the form of fluorapatite or carbonate fluorapatite. Phosphorite deposits tend to possess elevated uranium, thorium, rare earth element, yttrium, heavy metal and metalloid values [1 and 2]. Uranium and thorium concentrations of phosphate deposits

are highly variable and igneous deposits tend to contain lower uranium and higher thorium levels than sedimentary phosphorites [3 and 4]. Uranium content of phosphate rock varies from 20 ppm to 500 ppm with average concentration of 100 ppm in most phosphate rocks. The World Distribution of Uranium Deposits (UDEPO)

database tabulates 13.8 million tons of uranium in phosphate rock deposits through 2015 [5 and 6]. The discovery of phosphate rocks in Egypt dates to the end of the last century [7]. Phosphate deposits in Egypt are part of the Middle East to the North African phosphogenic province of the late Cretaceous-paleogene age [8]. Egypt phosphate deposits are extended to about 750 km from the Red Sea coast to the El-Dakhla oases, present in three localities, east-west trending facies belts, phosphorite northern facies belt, phosphorite of the central facies belt and phosphorite of the southern facies belt [8]. Phosphorite of the central facies belt represents the most economic occurrences which is confined into three localities, the Red Sea coast from Safaga to the Quseir land-stretch, the Nile Valley between Idfu and Qena and the western desert on the Abu Tartur plateau

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(New Valley area) [9 and 10]. According to The United Nations Framework Classification for Fossil Energy and Mineral Reserves and Resources 2009, quantities of phosphate rocks in the Nile Valley are 49.0 Mt proved reserves[11]. Potential issues of concern resulting from phosphate mining are its radiological impacts; possible increases in external exposure or internal exposure through direct ingestion or inhalation. From the natural risk point of view, it is necessary to know the dose limits of public exposures and to measure the natural environmental radiation level provided by phosphate ore. In this study a survey was carried out to evaluate the activity concentrations and assess the associated hazard indices and radiological doses of the naturally occurring radionuclides ^{226}Ra , ^{232}Th , and ^{40}K in phosphate samples from EL-Naser phosphate quarry in El-Mahamidarea, a part of Nile valley, which is considered the most important areas of presence of Phosphorite of the central facies belt [12, 13 and 14].

Materials and Methods

A number of phosphate soil samples each is 1Kg in weight were collected from different sites of crushing and grinding of phosphate rocks at El-Naser phosphate quarry in El-Mahamid area, Egypt. The samples were properly marked, catalogued and brought to the Laboratory for preparation before measuring. Samples with large grain size were crashed then they were dried. Afterwards, the samples were sieved to a fine grain size powder. Every powdered sample was mixed to obtain a homogeneous sample. The representative samples were weighted and carefully sealed in tight containers to avoid any possibility of out gassing and were stored for 4 weeks to reach equilibrium [15,16 and 17].

Low-level background gamma-ray spectrometer consists basically of 3×3 inch NaI(Tl), S-1212-I model, with a 1024 microcomputer multichannel analyzer, 5510 ORTEC Norl and housed in thick lead shield used to estimate the specific activity of samples under investigation. The applied detector has a peak gamma-ray efficiency of 1.2×10^{-5} at 1332 keV, energy resolution of 7.5 % at 662 KeV[18 and 19] and operation bias voltage 800-1000 V D.C. The detector was calibrated for various γ -ray energies by using ^{60}Co (1173.2 and 1332.5 KeV) and ^{137}Cs (661.64 KeV). Byconsidering the secular equilibrium, ^{238}U and ^{232}Th activities were determined via their daughters activities where ^{214}Bi (609.3, 1120.3 and 1764KeV) and ^{214}Pb (351 KeV) were used for determination the activity concentration of ^{238}U and ^{208}Tl (2614 KeV), ^{212}Pb (238KeV) and ^{228}Ac (911 KeV) were used fordetermination the activity concentration of ^{232}Th , while the activity concentration of ^{40}K were determined from its 1460(10.7%) KeV[18, 19, 20 and 21]. The background was measured and subtracted from the net count for all measured samples. The activity concentrations of the natural radionuclides in the measured samples were computed using the following equation [22 and

$$23]. A_s = \frac{N_C}{M} \times \frac{1}{\tau} \times \frac{1}{\eta} \text{ Bq/Kg (1)}$$

Where N_C is the net counting rate of γ -ray (counts per second) corrected for background η the detector efficiency of the specific γ -ray, τ the absolute transition probability of γ -decay and M the mass of the sample in Kg. The activity concentrations of ^{226}Ra , ^{232}Th can be expressed in terms of (ppm) and ^{40}K in (%) where the specific activity of a sample containing 1ppm by weight of ^{226}Ra is 12.35 Bq/Kg, ^{232}Th is 4.06 Bq/Kg and 1% of ^{40}K is 313Bq/Kg[15, 22, 23 and 24].



Location Map of The Study Area

Radium equivalent (Ra_{eq})

The radioactivity has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq/Kg to represent the activity levels of ^{226}Ra , ^{232}Th and ^{40}K by a single quantity by using the given equation [25]. $Ra_{eq} \text{ (Bq/Kg)} = A_{Ra} + 1.43A_{Th} + 0.077A_K$ (2)

Where A_{Ra} , A_{Th} and A_K are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K respectively.

The absorbed dose rates (D_{out})

The absorbed dose rate (D_{out}) 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 are calculated from the following formula [25 and 26].

$$D_{out} \text{ (nGy h}^{-1}\text{)} = 0.462A_{Ra} + 0.621A_{Th} + 0.0417A_K \quad (3)$$

Where 0.462, 0.621 and 0.0417 are conversion factors in nGy h^{-1} per unit activity concentration in Bq/Kg (dry weight) for ^{226}Ra , ^{232}Th and ^{40}K respectively.

The annual effective dose (D_{eff})

The conversion coefficient from absorbed dose in air to effective dose (0.7SvGy^{-1}) and outdoor occupancy factor (0.2) proposed by UNSCEAR [25, 26 and 27] are used to estimate the annual effective dose rates $D_{eff} \text{ (mSvyr}^{-1}\text{)}$. Therefore, the outdoor annual effective dose equivalent can be given as follow.

$$D_{eff} \text{ (mSvyr}^{-1}\text{)} = D \text{ (nGy h}^{-1}\text{)} \times 8760 \text{ h yr}^{-1} \times 0.7 \times (10^3 \text{ mSv/}10^9 \text{ nGy}) \times 0.2,$$

$$D_{eff} \text{ (mSvyr}^{-1}\text{)} = D \text{ (nGy h}^{-1}\text{)} \times 1.21 \times 10^{-3} \text{ (mSvyr}^{-1}\text{)} \quad (4)$$

External and Internal hazard Indices (H_{ex} & H_{in})

The external and internal hazard indices are defined as [25].

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \quad (5)$$

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \quad (6)$$

The values of H_{ex} and H_{in} must be less than unity to keep the radiation hazard insignificant.

Excess lifetime cancer risk (T_{cr})

The possibility of cancer development due to exposure to radiation, taking into account the average age of human 70 years was calculated

from the following relation

$$T_{cr} = D_{eff} \text{ (mSvyr}^{-1}\text{)} \times \phi_L \times R_f \quad (7)$$

Where ϕ_L is the average duration of a lifetime (estimated to be 70 yrs.) and R_f is the risk factor (Sv), for stochastic effects, assigned by ICRP as $0.05/\text{Sv}$ for the public [28 and 29].

Annual gonadal equivalent dose (D_{geff})

Measurement of the threat resulting from the effect of a certain level of radiation on the gonads called annual gonadal equivalent dose and calculated from the following relation [28, 29, 30 and 31]. $D_{geff} \text{ (mSvyr}^{-1}\text{)} = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \quad (8)$

Results and Discussions

The measured activity concentrations as well as the uncertainty of the naturally occurring radio nuclides ^{226}Ra , ^{232}Th and ^{40}K in Bq/Kg for phosphate soil samples from the inside border of El-Naser phosphate quarry in El-Mahamid area are listed in Table 1 and Fig. 1 where the activity concentration of ^{226}Ra ranged from 277.37 ± 9.09 to 590.90 ± 19 Bq/Kg with average value 452.06 ± 14.72 Bq/Kg, ^{232}Th ranged from 20.69 ± 0.7 to 55.12 ± 4.19 Bq/Kg with average value 49.34 ± 1.68 Bq/Kg and ^{40}K ranged from 244.44 ± 8.52 to 545.63 ± 18.80 Bq/Kg with average value 360.70 ± 12.49 Bq/Kg. From the obtained results it is clear that, the radium content in all samples is higher than ^{232}Th and ^{40}K . The observed average concentrations of ^{226}Ra and ^{232}Th in Bq/Kg are higher than the world average of 35 Bq/Kg for ^{226}Ra and 30 Bq/Kg for ^{232}Th and the average concentration is lower as compared with the world average value of 400 Bq/Kg for ^{40}K [27 and 32]. From Table 1 the average values in ppm for the three natural radio nuclides equal 36.60, 9.58 ppm and 1.15% for ^{226}Ra , ^{232}Th and ^{40}K respectively. The average concentrations values of the present work were compared with similar studies in Egypt and many parts of the world and a summary was listed in Table 2. From listed results, the average value of ^{226}Ra of the present work is higher than the average values of 287 Bq/Kg for Abu-Tartor, wet rock reported by Khater, A. E., 2001 [2], 419.13 Bq/Kg for Abu-Tartor reported by S. M. Darwish, S. M., 2015 [24] and 222.4 to 255.8 Bq/Kg for Safaga, 122.4 to 188.3 Bq/Kg for El-Quseir and 115.4 to 165.8 Bq/Kg

Kg for El-Hamrawein reported by Atta, E. R., 2016[42]. The average value of ^{226}Ra of this work is higher than the average values of 255 Bq/Kg for Algeria reported by Boumala, D., 2018[49], 355Bq/Kg for South Koreareported by Chang, B. U., 2008[46] and 393 for Sudan reported by Sam, A. K., 1995[45]. The average value of ^{232}Th of this work is higher than the values of 23.7 Bq/Kg for Abu-Tartor, wet rockreported by Khater A. E., 2001[2], 16 Bq/Kg for Abu-Zaabal reported by Diab, H. M., 2008[25] and 37 Bq/Kg for Abu-Zaabal plant reported by Hussein, E. M., 1994[40] and lower than the values of 329.4 Bq/Kg for Wadi El-Mashash reported by Adel Abbady, A. G.,2005[41] and 135.6 to 212.3 Bq/Kg for Safaga, 112.8 to 167.4 Bq/Kg for El-Quseir and 132.8 to 188.6 for El-Hamrawein reported by Atta, E. R., 2016[42]. The average value of ^{232}Th is much less than the values of 447.62 Bq/Kg for Tanzania reported by Meza, L. H., 2015[48] and 193 Bq/Kg for Algeria reported by Boumala, D., 2018[49], higher than the values of 2 Bq/Kg for Jordan reported by Olszewska-Wasiolek, M., 1995[44], 4 Bq/Kg for South Korea reported by Chang, B. U., 2008[46], 6.9 Bq/Kg for Sudan reported by Sam, A. K., 1995[45] and matches with those for Tunisia reported by Olszewska-Wasiolek, M., 1995 [44], Pakistan reported by Tufail, M., 2010[47] and Algeria reported by Olszewska-Wasiolek, M., 1995[44]. The average value of ^{40}K is higher than those reported in Egypt and reported in other countries except the values of 1582 Bq/Kg for raw material of Abu-Zaabal, Egypt reported by Diab, H. M., 2008[25] and 587.6 Bq/Kg for Wadi El-Mashash, eastern desert, Egypt reported by Adel Abbady, G.E., 2005[41].

From results listed in Table 3 the calculated values of radium equivalent Ra_{eq} ranged from 326 to 812 Bq/Kg with average value 550 Bq/Kg. The obtained results indicate that the values of radium equivalent activity for the most samples are above the the maximum value of 370 Bq/Kg which recommended by UNSCEAR[27, 33, 34 and 35]. From Table 3, the calculated values of absorbed gamma dose rate D_{out} varied from 157 to 321 nGyhr $^{-1}$ with average value 255 nGyhr $^{-1}$ is higher than the world average value of 55 nGyhr $^{-1}$ which recommended by UNSCEAR [27 and 35]. From the estimated values listed in *Egypt. J. Phy.* **Vol. 49** (2021)

Table 3, the annual effective dose D_{eff} varied from 0.19 to 0.39 mSvyr $^{-1}$ with average value 0.31 mSvyr $^{-1}$ is much higher than the corresponding world average value of 0.07 mSvyr $^{-1}$ for outdoor exposure[35 and 36].

The calculated values of external hazard index H_{ex} and internal hazard index H_{in} that represented in Table 3 ranged from 0.91 to 1.89 with an average value of 1.49 and from 1.66 to 3.47 with an average value of 2.71 for external hazard index H_{ex} and internal hazard index H_{in} respectively. The obtained results show that the externalhazard index H_{ex} and the internal hazard index H_{in} for all samples are higher than the maximum world value of unity [37 and 38].

The calculated values of the average excess lifetime cancer risk of the samples from inside the quarry also listed in Table 3 where the values ranged from 0.66 to 1.36 with average value 1.08 is greater than the world average of 0.29×10^{-3} [38]. The obtained values of annual gonadal dose equivalent listed in Table 3, the values ranged from 1057.85 to 2161.08 μSvyr^{-1} with average value 1672.70 mSvyr $^{-1}$ is exceed the average world allowed limit of 300 μSvyr^{-1} [39].

The calculated linear Pearson's correlation coefficients among ^{226}Ra and ^{232}Th , ^{232}Th and ^{40}K , and ^{226}Ra and ^{40}K and among the other radiological parameters using Statistical Program for Social Science (SPSS 25.0) are presented in Figs. 2, 3, 4 and in Table 4 for the studied samples. From obtained results, there is a very strong positive correlation ($r=0.752$) between ^{226}Ra and ^{40}K , while a strong correlation is observed between ^{226}Ra and ^{232}Th ($r=0.696$) and between ^{40}K and ^{232}Th ($r= 0.564$). The measured radioactive parameters are very strongly correlated with one another, and very strongly positively correlated with ^{226}Ra , ^{232}Th and ^{40}K .

Conclusion

The naturally occurring radionuclide ^{226}Ra , ^{232}Th , and ^{40}K have been measured in phosphate samples using NaI (TI) spectrometer and the values were compared with those reported in Egypt and other countries. The measured values of ^{226}Ra , ^{232}Th and ^{40}K were higher than the average world values recommended by UNSCEAR [27]. Furthermore, the hazard indices

and the doses rate associated were estimated. The radium equivalent (Ra_{eq}), external and internal hazard indices, the absorbed dose rates (D_{out}), the annual effective dose (D_{eff}), excess lifetime cancer risk (T_{cr}) and annual gonadal equivalent dose (D_{geff}) all were found to be higher than the allowed values given by UNSCEAR [27] which indicates the unhealthy impacts on humans and environment which requires an additional radiological attention for protection. This study has established information regarded to the background radiation levels and figured out the baseline data of the radioactivity levels related to phosphate activities which could utilizes as a reference for future studies.

Recommendations

From the current study, phosphate mining has negative impacts on humans and the environment if it is not been managed in a suitable way. Minimizing the negative impacts requires certain precautions in dealing with phosphate mining. From the author point of view to minimize these impacts the following recommendations are to be carried out :

1. The volatile dust in the quarry and its surrounding area can be treated by using more efficient dust collectors.
2. A full investigation and air quality monitoring nearby the mining area and conduct periodical medical surveys of the workers are highly recommended.
3. Do not expand the establishment of residential communities nearby the mining area.

TABLE 1. The Activity Concentration of ^{226}Ra , ^{232}Th and ^{40}K , ^{226}Ra , ^{232}Th Content and ^{40}K Percentage for Phosphate Samples

Sample NO.	Activity Concentration Bq/Kg					
	^{226}Ra	^{232}Th	^{40}K	^{226}Ra (ppm)	^{232}Th (ppm)	$^{40}\text{K}\%$
P1	590.90 ± 19.18	51.88 ± 1.76	376.880 ± 13.05	47.85	12.78	1.20
P2	554.52 ± 18.05	35.13 ± 1.21	545.630 ± 18.80	44.90	8.65	1.74
P3	483.33 ± 15.72	55.00 ± 4.19	481.640 ± 16.58	39.14	13.55	1.54
P4	453.46 ± 14.79	45.00 ± 2.34	351.420 ± 12.21	36.72	11.08	1.12
P5	524.20 ± 17.03	46.23 ± 1.57	394.100 ± 13.62	42.45	11.39	1.26
P6	382.18 ± 12.46	31.05 ± 1.06	244.440 ± 8.52	30.95	7.65	0.78
P7	425.11 ± 13.86	36.71 ± 1.26	314.330 ± 10.92	34.42	9.04	1.00
P8	377.42 ± 12.30	20.69 ± 0.71	276.360 ± 9.60	30.56	5.10	0.88
P9	277.37 ± 9.09	28.39 ± 0.98	261.500 ± 9.09	22.46	6.99	0.84
Average	452.06 ± 14.72	49.34 ± 1.68	360.70 ± 12.49	36.60	9.58	1.15
Max	590.90 ± 19	55.00 ± 4.19	545.63 ± 18.80	47.85	13.55	1.74
Min	277.37 ± 9.09	20.69 ± 0.7	244.44 ± 8.52	22.46	5.10	0.78

TABLE 2 . Comparison of the Average Values of the Activity Concentration in Bq/Kg of ^{226}Ra , ^{232}Th and ^{40}K of Present Study with Similar in Egypt and Different Countries.

Country	Activity concentration (Bq/Kg)			References
	^{226}Ra	^{232}Th	^{40}K	
Present work	452.06	49.34	360.70	--
Abu-Zaabal plant, Egypt	514	37	19	40
Abu-Tartor, wet rock, Egypt	287	23.7	21.4	2
Wadi El-Mashash, Egypt	665.8	329.4	587.6	41
Abu-Zaabal, Egypt	1180.6	16	1582	25
Wadi Qena, Egypt	864.69	54.14	87.39	24
Abu-Tartor, Egypt	419.13	48.21	115.95	24
Safaga, Egypt	222.4 to 255.8	135.6 to 212.3	225.2 to 312.8	42
El-Quseir, Egypt	122.4 to 188.3	112.8 to 167.4	168.7 to 268.9	42
El-Hamrawein, Egypt	115.4 to 165.8	132.8 to 188.6	95.2 to 155.8	42
Egypt	571	19	182	43
Jordan	1044	2	8	44
Tunisia	821	29	32	44
Algeria	619	64	22	44
Sudan	393	6.9	141	45
South Korea	355	4	49	46
Pakistan	511	52	206	47
Tanzania	1832.32	447.62	399.46	48
Algeria	255	193	23	49

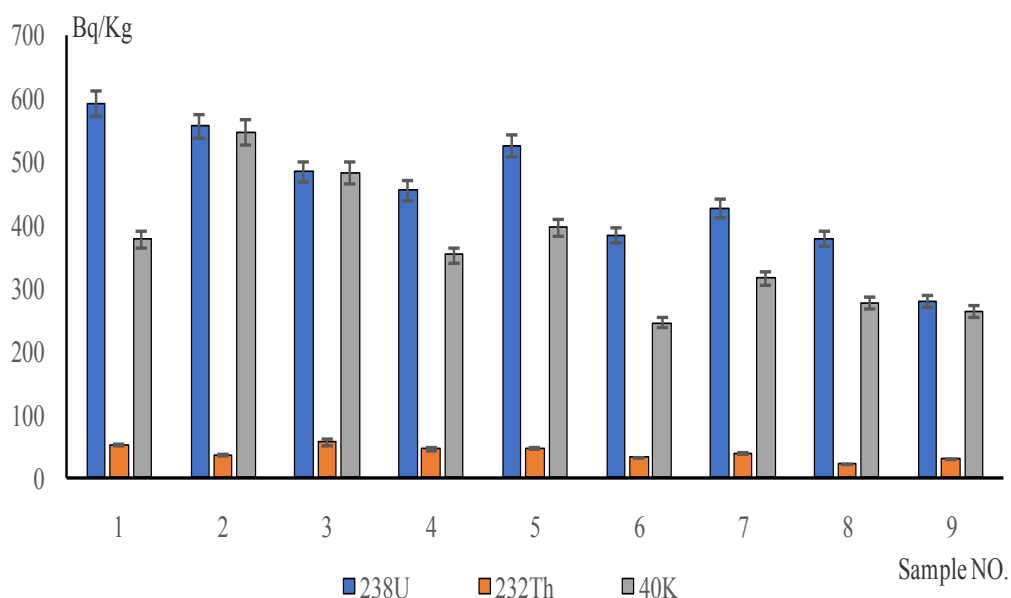
**Fig. 1. ^{226}Ra , ^{232}Th and ^{40}K Activity Concentrations for all Phosphate Samples.**

TABLE 3. Radiation Hazard Indices and Radiation Doses for Phosphate Samples.

Sample NO	R_{eq} (Bq/kg)	D_{out} (nGyh ⁻¹)	D_{eff} (mSvy ⁻¹)	H_{ex}	H_{in}	(Tcr)	D_{geff} (μSvy ⁻¹)
P1	694.12	320.93	0.39	1.88	3.47	1.36	2161.08
P2	646.76	300.75	0.36	1.75	3.25	1.27	2031.64
P3	699.34	321.08	0.39	1.89	3.20	1.36	1874.62
P4	578.98	266.91	0.32	1.56	2.79	1.13	1699.64
P5	620.66	287.33	0.35	1.68	3.09	1.22	1936.77
P6	445.40	206.04	0.25	1.20	2.24	0.87	1387.48
P7	501.81	232.31	0.28	1.36	2.50	0.98	1565.74
P8	428.29	198.74	0.24	1.16	2.18	0.84	1339.49
P9	338.10	156.68	0.19	0.91	1.66	0.66	1057.85
Average	550.39	254.53	0.31	1.49	2.71	1.08	1672.70
Max	699.34	321.08	0.39	1.89	3.47	1.36	2161.08
Min	338.10	156.68	0.19	0.91	1.66	0.66	1057.85

TABLE 4. Pearson Correlation Coefficients Between Radioactive Parameters for Phosphate Samples.

	²²⁶ Ra	²³² Th	⁴⁰ K	Ra_{eq}	D_{out}	D_{eff}	H_{ex}	H_{in}	D_{geff}	T_{cr}
²²⁶ Ra	1									
²³² Th	.696*	1								
⁴⁰ K	.752*	0.564	1							
Ra_{eq}	.935**	.853**	.825**	1						
D_{out}	.941**	.846**	.829**	1.000**	1					
D_{eff}	.939**	.854**	.819**	1.000**	1.000**	1				
H_{ex}	.937**	.851**	.825**	1.000**	1.000**	1.000**	1			
H_{in}	.979**	.796*	.808**	.988**	.990**	.989**	.989**	1		
D_{geff}	.993**	.763*	.791*	.966**	.970**	.969**	.968**	.994**	1	
T_{cr}	.941**	.848**	.826**	1.000**	1.000**	1.000**	1.000**	.990**	.970**	1

**. Correlation is significant at the 0.01 level.

*. Correlation is significant at the 0.05 level.

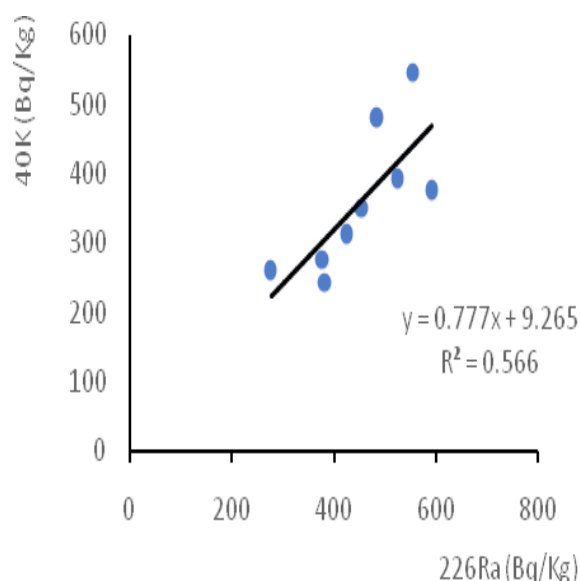


Fig. 2. Correlation Between ^{226}Ra and ^{40}K for all Phosphate Samples.

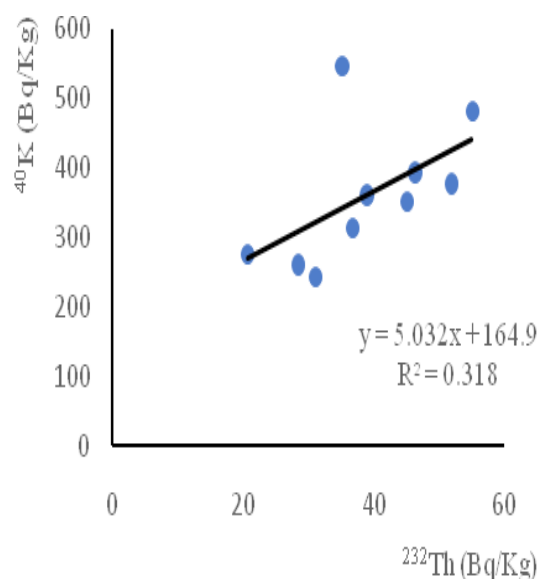


Fig. 3. Correlation Between ^{232}Th and ^{40}K for all Phosphate Samples.

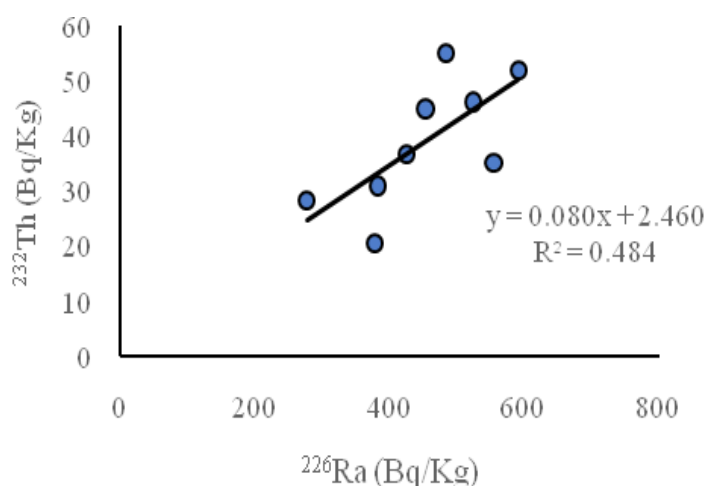


Fig. 4. Correlation Between ^{226}Ra and ^{232}Th for all Phosphate Samples.

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