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Indoor Radon Concentration for Phosphate Rocks Samples Using CR-39 Detector.

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A B S T R A C T

Radon concentrations and exhalation rate were measured using Can Technique with CR-39 plastic track detectors for phosphate rocks samples from Safaga and El-Hamrawayn areas in the Eastern Desert, Egypt. The values of radon concentrations ranged from 1362.21- 20045.30 Bqm^{-3}, 764.84 -17828.48 Bqm^{-3} and the values of surface exhalation rate ranged from 1.22-18.07 $Bqm^{-2}h^{-1}$, *1.09-16.07* $Bqm^{-2}h^{-1}$ for Safaga and El-Hamrawayn, respectively. From the results we can conclude that the values of radon concentration in Safaga higher than El-Hamrawayn. The present study aimed to detect any harmful radiation that would affect the human and radioactivity background levels.

Introduction:

Radon is produced through α-decay of ^{226}Ra in the soil is the only gaseous decay in this series as a noble gas, part of the ^{222}Rn emanates from the soil grains into the air and diffuse to the atmosphere. Radon is the most important natural radioactive factor harmfully the human population, because radon is radioactive gas comes from the natural decay of uranium deposits in soil, rocks and water (Swakon et al., 2005).

The largest phosphate rock deposits worldwide are located in one belt covering all North African countries and continue through Jordan and North West of Saudi Arabia (Ragheb and Khasawneh, 2010). The Egyptian phosphate is widely distributed in many locatities on the Red Sea, Nile Valley and Western Desert. Phosphate rock is the starting raw material for all phosphate products and its decay products tend to be elevated in phosphate deposits of sedimentary origin (Ashraf et al., 2001).

The use of phosphate fertilizers in the recent years leads to accumulation of ^{238}U and ^{232}Th in soil, which cause pollution soil, drainage water and transmitted to animals and eventually to man who consumes the meat or milk from those animals. Most of the heavy metals accumulate in the food chains of the ecosystem (El-Zakla et al., 2007). Fertilizers are used for reclaiming the land and improving the properties of crops like super phosphate which, most commonly used in Egypt. It is manufactured from the reaction between sulfuric acid, phosphate rock and water (Ashraf et al., 2004; Rehman et al., 2006). Phosphate deposit of sedimentary origin contains higher concentration of ^{238}U and its decay products than phosphate from volcanic or biological origin (Korkmaz et al., 2005). The investigated samples were collected from Safaga and El-Hamrawayn areas of the Eastern Desert along the Red Sea. The phosphate deposits of the Eastern Desert along the beach of Red Sea from Safaga to Quseir between latitude (25° 00' - 26° 47') and longitudes (33° 45' - 34° 25').

The present work is aiming to determine the radon concentrations and radon exhalation rate in the phosphate rocks samples from Safaga and El-Hamrawayn areas in the Eastern Desert along the beach of Red Sea. In order to detect any harmful radiation that would affect the human and radioactivity background level this, can be used as reference information to assess any changes in the radioactive background level in order to detect any harmful radiation that would affect the human and the environment.

Materials and Methods:

Twenty phosphate rocks samples were measured using Can technique to determine the values of radon concentration and exhalation rate with CR-39 detector. Ten samples were collected from Safaga and the other ten samples were collected from El-Hamrawayn. The samples were collected at equal distances 500 *m* from each other. The samples were crushed, dried in oven at

110°*C* for 3hr, minced, sieved by 1-*mm* mesh and weighted. The samples were carefully sealed for thirty days in plastic cylindrical containers with dimensions of 6 *cm* in diameter and 12 *cm* in depth. Each sample container was capped tightly to an inverted cylindrical plastic cover as shown in figure 1.

A piece of CR-39 of 700 *µm* thickness (American Technical Plastic, Inc.) detector of area (1.5 x 1.5) cm^2 fixed at the bottom center of the inverted plastic cover. After the irradiation period, the detectors were collected and chemically etched in NaOH solution 6.25N at 70°*C* for 7 *hr* **(**Yip et al., 2003**)**. After etching the CR-39 detectors were washed in distilled water and then dipped for few minutes in a 3 % acetic acid solution and washed again with distilled water and finally air dried.

The track density was determined by using optical microscope **(**Hafez et al., 2011**)** which calibrated before usages. The background of CR-39 track detector was counted by optical microscope and subtracted from the count of all detectors **(**Abo-Elmagd et al., 2006**).** The value of radon concentration in (Bqm^{-3}) at secular equilibrium is given by the following equation:

$$C_{Rn} = \frac{\rho}{\eta T} \qquad (1)$$

Where, C_{Rn} is radon concentration (Bqm^{-3}), ρ is the track density *(track* cm^{-2}*)*, *T* is the exposure time *(day)*, and η is the calibration coefficient of CR-39 nuclear track detectors obtained from the experimental calibration 0.22 *tracks* $cm^{-2}day^{-1}/Bqm^{-3}$ of radon, respectively **(**Hafez et al., 2011**)**. Radon exhalation rate is given by the relation**:**

$$E_A = \frac{CV\lambda}{A[T + \frac{1}{\lambda}(e^{-\lambda T} - 1)]} \qquad (2)$$

Where, E_A is the surface exhalation rate in $(Bqm^{-2}h^{-1})$, *C* is the integrated radon exposure in $(Bqm^{-3}h)$, λ is the decay constant of radon (h^{-1}), *V* is the effective volume of the cup (m^3), *A* is the cross section area of the can (m^2) and *T* is the exposure time (Barooah et al., 2011).

$$E_M = \frac{CV\lambda}{M[T + \frac{1}{\lambda}(e^{-\lambda T} - 1)]} \qquad (3)$$

Where, E_M is the mass exhalation rate in $(Bqkg^{-1}h^{-1})$ and *M* is the mass of the sample (Barooah et al., 2011). The effective radium content of the sample can be calculated using the formula:

$$C_{Ra} = \frac{\rho h A}{\eta T_e M} \qquad (4)$$

Where, *M* is the mass of the sample in *(kg)*, *A* is the area of cross section of the can in (m^2), η *h* is the distance between the detector and top of the sample in *(m)* (Mohamed, 2012). The annual absorbed dose rate (D_{Rn}) in $mSvy^{-1}$ due to the inhalation of radon in indoor air, was calculated according to the following equation (UNSCEAR, 2000).

$$D_{Rn}\ (mSvy^{-1}) = C_{Rn} \,.\, D \,.\, F \,.\, T \,.\, H \qquad (5)$$

Where, C_{Rn} is the measured mean radon concentration in air, *F* (0.4) is the indoor equilibrium factor. *T* is the indoor occupancy time *(hr)*, *H* is the indoor occupancy factor (0.4) and *D* is the dose conversion factor $(9\ x10^{-6}\ mSv\ h^{-1}/\ Bqm^{-3})$ (UNSCEAR, 2000; Maged, 2006; Nsiah et al., 2011).

The working levels were calculated using the following equation:

$$WL = \frac{C.F}{3700} \qquad (6)$$

Where, C_{Rn} is radon concentration in Bqm^{-3} and *F* is the equilibrium factor for radon has been taken as 0.4 (Mamta et al., 2011).

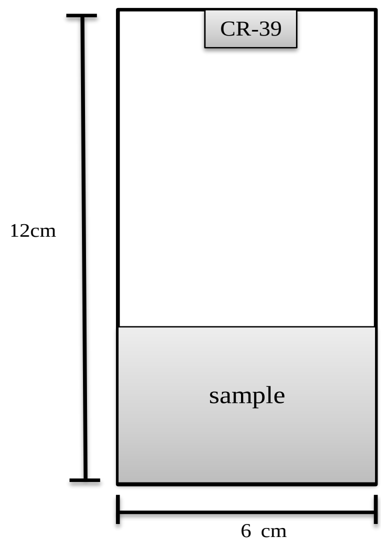

Fig: 1. Plastic cylindrical container

Results and Discussion:

The values of radon concentrations depend on many physical properties of the sample, like the chemical composition, porosity and bulk density of the samples. The values of radon concentration were measured, exhalation rate, radium concentration, annual absorbed dose rate and working level for the phosphate samples using CR-39 calculated as the following in table 1. The values of radon concentration ranged from 1362.21-20045.30 Bqm^{-3}, surface exhalation rate ranged from 1.22-18.07 $Bqm^{-2}h^{-1}$, mass exhalation rate ranged from 21.06 - 294.40 $mBqkg^{-1}h^{-1}$, radium concentration ranged from 2.39-32.00 $Bqkg^{-1}$,annual absorbed dose rate ranged from 17.10- 252 $mSvy^{-1}$ and the working level ranged from 2167.70 - 147.26 *m*WL for Safaga area are listed in table 1.

But in El-Hamrawayn the values of radon concentration ranged from 764.84-17828.48 Bqm^{-3}, surface exhalation rate from 1.09-16.07 $Bqm^{-2}h^{-1}$, mass exhalation rate from 27.14 -280 $mBqkg^{-1}h^{-1}$, radium concentration from 2.78-29.99 $Bqkg^{-1}$, annual absorbed dose rate from 9.64-244 $mSvy^{-1}$ and the working level from 82.26 to 1927.40 *m*WL. Figure 2, shows the comparison between the values of radon concentration for the studied areas. From the figure the average values of radon concentration in safaga higher than El-Hamrawayn and also the average surface exhalation rate in safaga higher than El-Hamrawayn as shown in figure 3. The comparison between the annual absorbed dose rate for safaga and El-Hamrawayn given by figure 4. The correlation coefficient between radon concentration and surface exhalation rate is $R^2 = 0.98$ as shown figure 5, this is very good relation to evaluate the radon risk in atmosphere. Assessing relation between radon an exhalation rate is necessary for the studied samples. Figure 6, shows the correlation relation between radium concentration and radon concentration for safaga area which, equal 0.92, this result shows a good linear relationship. Also the correlation relation between radon concentration and surface exhalation rate(R^2 = 0.87) and the correlation relation between radium concentration and radon concentration(R^2 = 0.90) as shown in figure 7 and figure 8 for El-Hamrawayn, respectively.

It seems that the Hamrawayn deposit has the lowest radioactivity level of exploited phosphate of sedimentary origin. The comparison between the obtained experimental results and the published data in different countries given by table 2.

Table: 1. Radon concentration (C_{Rn}), surface exhalation rate (E_A), mass exhalation rate (E_m), radium concentration (C_{Ra}) annual absorbed dose rate (D) and working level (WL) for the phosphate samples

Areas	No.	C_{Rn} (Bqm^{-3})	E_A $(Bqm^{-2}h^{-1})$	E_m $(mBqkg^{-1}h^{-1})$	C_{Ra} $(Bqkg^{-1})$	D$(mSvy^{-1})$	*m*WL
Safaga	1	8338.93 ± 95.62	7.51 ± 0.08	135.00 ± 1.54	14.68 ± 0.07	105.00	901.50
	2	2836.97 ± 46.20	4.07 ± 0.06	94.88 ± 1.55	9.55 ± 0.03	35.78	306.69
	3	2160.49 ± 48,67	3.10 ± 0.06	69.09 ± 1.35	7.07 ± 0.02	32.99	233.56
	4	4946.66 ± 73.57	4.45 ± 0.6	72.30 ± 1.07	7.86 ± 0.05	62.38	534.77
	5	20045.30 ± 152.08	18.07 ± 0.13	294.40 ± 2.23	32.00 ± 0.10	252.00	2167.70
	6	4580.75 ± 70.80	6.57 ± 0.06	138.20 ± 1.07	14.15 ± 0.05	57.78	495.21
	7	12040.30 ± 114.62	10.85 ± 0.16	191.40 ± 3.50	20.80 ± 0.05	151.02	1301.65
	8	7006.97 ± 87.56	6.31 ± 0.07	99.50 ± 1.24	10.81 ± 0.08	88.38	757.51
	9	19278.18 ± 145.23	17.38 ± 0.13	232.00 ± 2.20	31.85 ± 0.10	243.11	2084.12
	10	1362.21 ± 38.60	1.22 ± 0.12	21.60 ± 0.61	2.35 ± 0.02	17.10	147.26
El Hamrawayn	1	12660.41 ± 117.69	11.41 ± 0.10	190.00 ± 1.77	20.70 ± 0.08	159.01	1368.69
	2	17613.94 ± 138.82	15.89 ± 0.12	280.00 ± 2.20	20.86 ± 0.10	222.03	1904.20
	3	764.84 ± 28.62	1.09 ± 0.04	27.14 ± 1.13	2.78 ± 0.01	9.64	82.26
	4	9729.21 ± 103.17	8.77 ± 0.09	148.50 ± 1.57	16.14 ± 0.07	122.72	1051.80
	5	4525.60 ± 70,36	6.50 ± 0.10	188.20 ± 2.95	19.46 ± 0.04	56.52	484.25
	6	17828.48 ± 139.66	16.07 ± 0.12	275.80 ± 2.16	29.99 ± 0.10	244.00	1927.40
	7	1086.51 ± 34.37	1.56 ± 0.04	36.11 ± 1.14	3.69 ± 0.02	13.71	117.46
	8	6084.45 ± 81.59	5.48 ± 0.07	83.00 ± 1.11	9.02 ± 0.05	76.01	657.77
	9	10590 .43 ± 107.64	9.54 ± 0.09	152.30 ± 1.54	16.50 ± 0.07	133.00	1144.91
	10	14552.00 ± 126.00	7.51 ± 0.11	253.40 ± 2.19	27.51 ± 0.23	183.50	1573.23

Table: 2. The comparison between the obtained results and the published data in different countries

Country	C_{Rn} (Bqm^{-3})	E_A $(Bqm^{-2}h^{-1})$	C_{Ra} $(Bqkg^{-1})$	References
Egypt	8.90 - 1675.40	0.02 - 4.12		(Saad, 2008)
Egypt	10.00 - 1244.00	0.05 - 2.35		(Maged et al., 1998)
Iraq	13.00 - 1197.00	o.26 - 2.40	0.04 - 6.78	(Jebur et al., 2014)
Iraq	72.20 - 177.50			(Karim et al., 2011)
Saudi	17.00 - 77.00	0.12 - 0.57		(Fatimh, 2014)
Saudi		0.04 - 0.35	4.27 - 33.93	(Kadi et al., 2011)
Iran	107.92 - 470.18		9.10 - 16.40	(Kant et al., 2006)
Nigeria	36.60 - 117.00			(Okeji et al., 2012)
Egypt:(Safaga)	1362.21-20045.30	1.22-18.07	2.39-32.00	The present work
(Hamrawayn)	764.84-17828.48	1.09-16.07	2.78-29.99	The present work

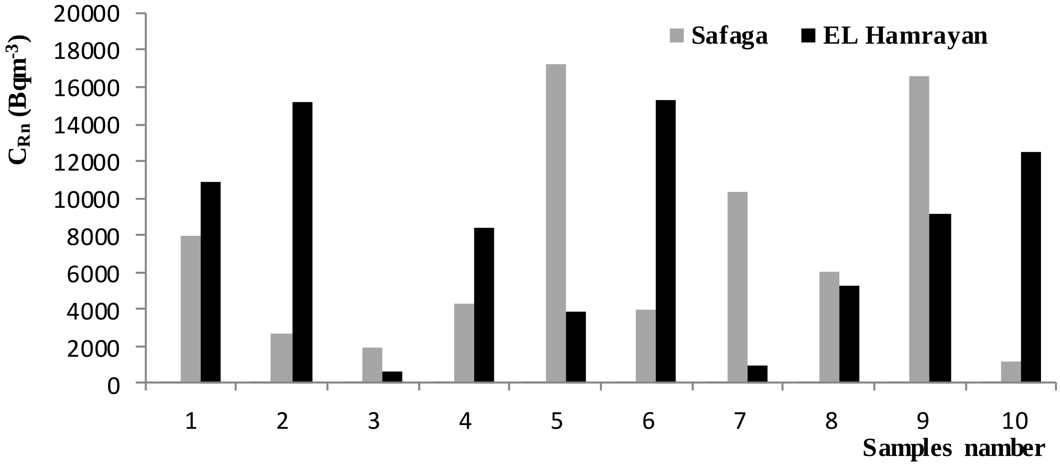

Fig: 2. The comparison between radon concentrations of phosphate samples for Safaga and EL Hamrayan areas

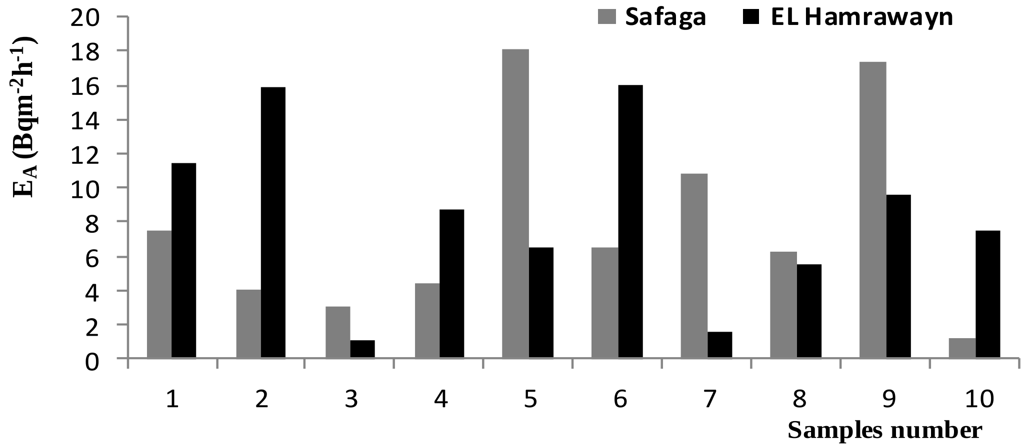

Fig: 3. The comparison between the surface exhalation rate of phosphate samples for Safaga and EL Hamrawayn areas

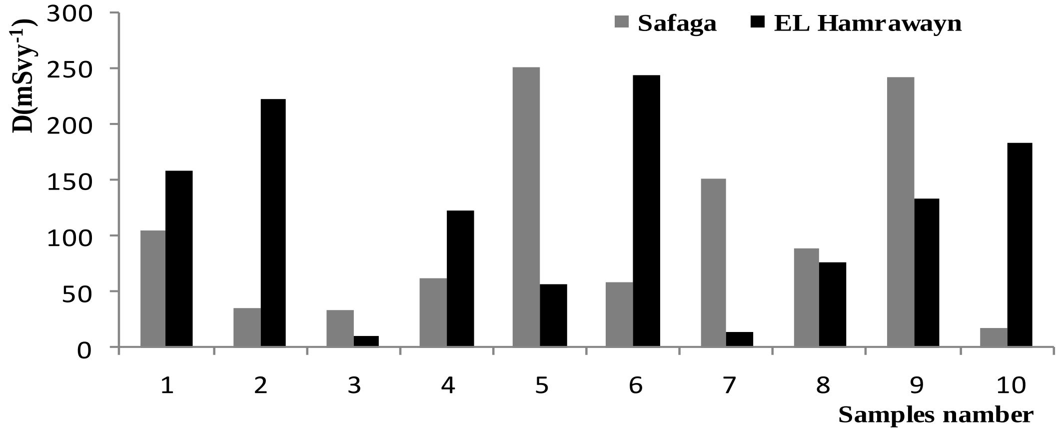

Fig: 4. The comparison between the annual absorded dose of phsphate samples for Safaga and EL Hamrawayn areas

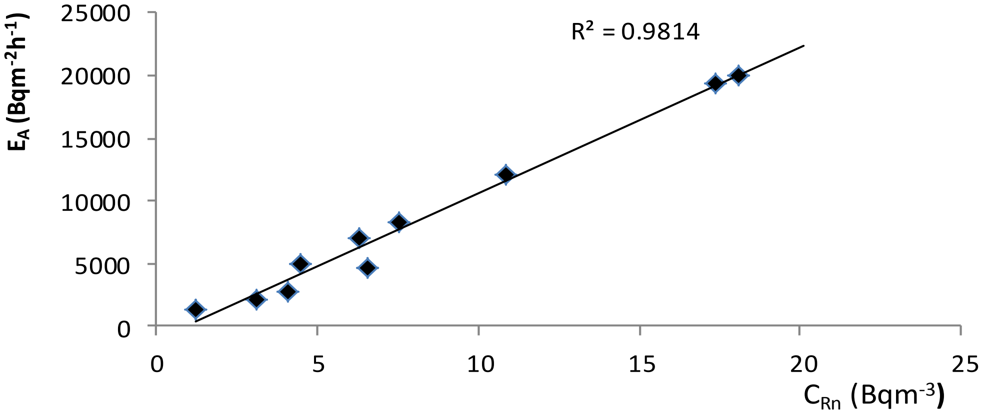

Fig: 5. The correlation between radon concentration(C_{Rn}) and surface exhalation rate (E_A) for samples in Safaga area

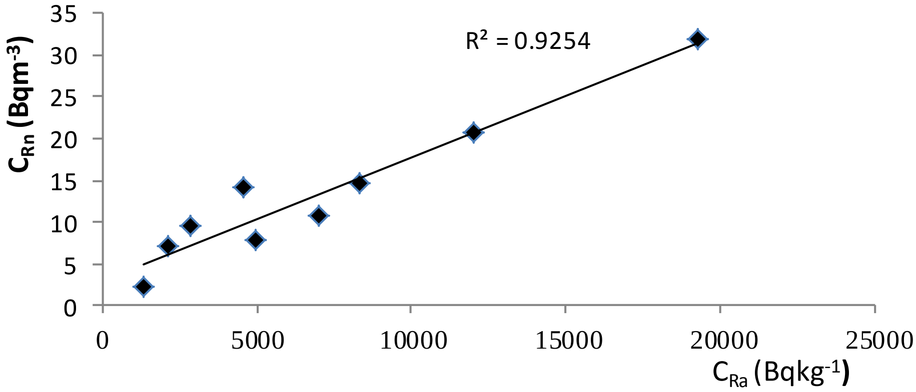

Fig: 6. The Correlation bet ween radon concentration(C_{Rn}) and Radium content(C_{Ra}) for the samples in Safaga area

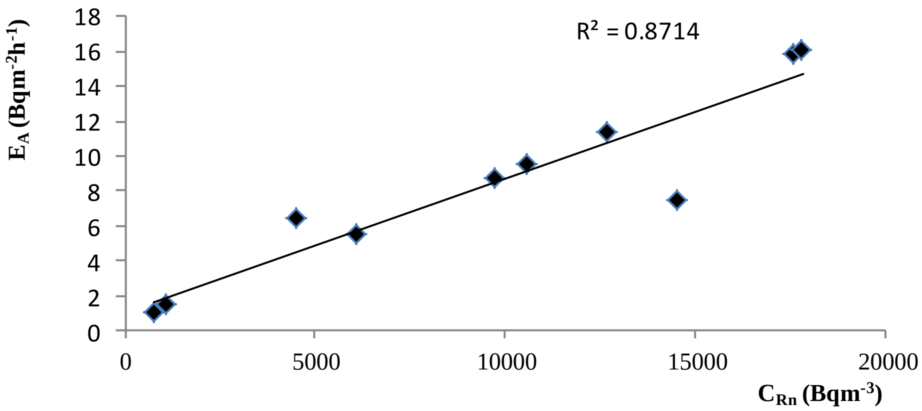

Fig: 7. The correlation between radon concentration(C_{Rn}) and exhalation rate(E_A) for the samples in El-Hamrawayn area

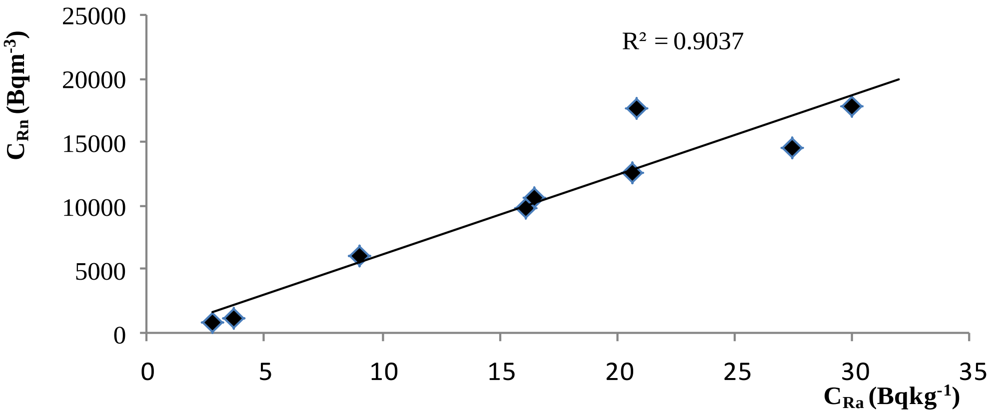

Fig: 8. The correlation between radon concentration (C_{Rn}) and radium concentration (C_{Ra}) for the samples of El Hamrawayn area

Conclusion:

This work is important to detect any harmful radiation that would affect the human and radioactivity background level which, can be used as reference information to assess any changes in the radioactive background level. Regarding phosphate fertilizers, the problem of radioactivity is raised at two levels, at the storage level where fertilizers continue to exhale ^{222}Rn in the surrounding atmosphere. The level of fertilizer use where radioactive elements are thought to get into plants and the passage of radionuclides in the food chain starts with phosphate fertilizer (IAEA, 2003).

The values of radon concentrations ranged from 1362.21-20045.30 *Bqm*$^{-3}$, 764.84-17828.48 *Bqm*$^{-3}$ for Safaga and El-Hamrawayn areas, respectively. The variations in the values of radon concentrations due to the difference in the chemical composition and the geological form of the samples. The values higher than the range of action levels from 200 to 600 *Bqm*$^{-3}$ recommended by (ICRP, 1994).

The average dose received by the workers of the phosphate mine are 0.15 to 0.29 *mSvy*$^{-1}$, which is far below the permitted dose of 20 *mSvy*$^{-1}$ recommended by the (ICRP Publication 60, 1990). The maximum permissible safe radiation dose without harm to the individual*(1 mSvy*$^{-1}$*)*, with continuous external irradiation of the whole body.

We conclude that, the values of radon concentrations in the phosphate are higher than the worldwide limit and not safety for human. The exposure and dose rates exceeded public permissible values in the sedimentary phosphate rocks, so that we must use personal protective masks to protect ourselves from inhalation alpha particles and don't live near the area under study to minimize the exposure time of radiation. So that we must repeat the measurements to detect the variation in the concentration of radioactive radionuclides that effect in the environment and human.

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