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Radium Content and Radon Exhalation Rate from Natural Samples Using SSNTD

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Abstract: Radium levels in some soil and fertilizers are analyzed from radiation protection perspective. In this work, evaluation of Radon concentration, radium activity, the radon exhalation rate in some fertilizers and soil sample in Khabat village Sohage governorate, Egypt, were addressed and discussed using CR-39 nuclear track detector. In that technique, radon gas passively diffused into the detector. For radium measurements, CR-39 Samples have been calibrated by using some natural samples with known radium activity. The radium activity in the natural sample has been determined using high pure germanium detector. Linear relationship between track density and radium activity has been achieved. It is found that the calibration factor of CR-39 for radium measurements is (0.014 Trcks.cm^{-2}.day/Bq.kg^{-1}). The mass and areal Radon exhalation rate show linear relation with radium content in the samples .The radon gas concentration show large variation with the maximum concentration ((7523.81 Bq/m^3) in the waste product of Abo-Zaboul factory. The minimum value (71.43Bq/m^3) is found in ammonium chemical fertilizer sample.Values of radium content in all soil samples except those near Abo-Zaboul factory are less than the permitted value of 370 Bq/kg authorized by organization for Economic corporation and development.

Keywords: Radium activity, Radon, fertilizers, CR-39detector, Radonexhalation.

1 Introduction

It is known that, a major part of the total dose received from inhalation of Ra-226 decay product. Phosphate fertilizer can contribute to radium content in the soil in a limited way. The determination of radium isotopes is important not only in environmental studies but also for the protection of public health. Radium has four naturally occurring isotopes [1]. Three of the isotopes (^{226}Ra, ^{224}Ra and ^{223}Ra) disintegrate into radon isotopes [2]. A fertilizer from rock origins like phosphate contains some radioactive element in natural content so it is important to study the concentration of these elements in the fertilizers to evaluate the potential radiation hazard in the environment [3].Naturally occurring radionuclide are exist in the fertilizers. The most important radionuclide is ^{238}U decay series, specially the isotope of radium. The radionuclide concentration in the fertilizers is largely depends on the raw material that used in the manufacture of fertilizers. It has been known that phosphate ores contains considerable amount of uranium, thorium and radium and natural isotope of potassium ^{40}K [4,5].^{222}Rn is a member of ^{238}U series and a decay product of ^{226}Ra, the radon concentration in the natural samples is dependent on the radium concentration in the ground which in turn dependent of uranium concentration in the soil. The rate at which radon released from ground into its surrounding environment is denoted as radon exhalation rate [6],determining its value is helpful to investigate the radon hazard on public health[7,8]. Radon exhalation is affected by many factors; the most important of them is the radium concentration in the soil [9]. As radon gas is inhaled its decay product is attached to inner surface of respiratory system and act as internal source of radiation cause large damage to cells that line it which may lead to lung cancer. It is known that, there is a strong relationship between radium activity and radon concentration anywhere [10]. Using phosphate fertilizers may be increase the concentration of radionuclide in soil with time [11]. The objective of this study was to evaluate the radium content and measure the radon activity concentration and exhalation rate in term of both mass and area for different samples of phosphate fertilizers using solid state nuclear

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track detector.

2 Experimental Sections

In this environmental study, a CR-39 track detector was used to evaluate the radon concentration and effective radium content. Samples of different types of fertilizers used in Egyptian market are collected.Also, a number of 22 soil samples were collected from El khabat village, which lies in the western south of Sohag governorate. Other samples were collected from Abo-Zaboul fertilizers phosphate factory, which lies in North West of Cairo. The samples were gathered at 10 cm depth from each point of sampling to eliminate organic waste martial from being contained within the sample. The samples are dried in the oven for one day at 110 C° then mashed and sieved and each 500 g of the soil samples are placed in plastic cylindrical container. Can of dimension 26 cm in height and 15 cm in diameter has been used. A piece of CR-39 detector of thickness 500µm and of area 1cm ×1cm is placed inside a plastic cup of 4cm in diameter and 10 cm in height and covered with plastic filter that fixed at the top of the container such that 23cm is the height of air between surface of sample and active side of the detector as shown in figure .1(a).

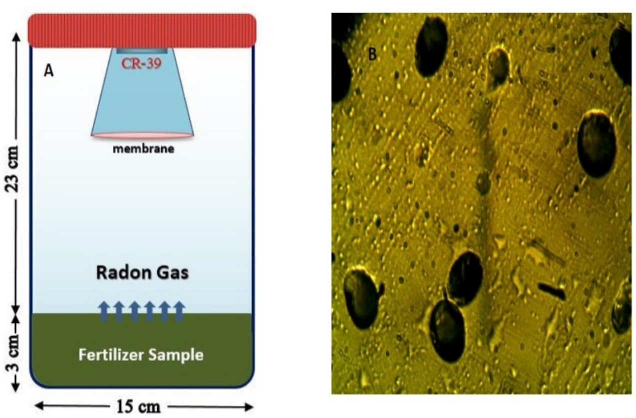

Fig.1(a). The plastic can used as radon irradiator.(B) typical image for alpha particles tracks.

The container is tightly sealed using silicon and plastic tape and left for 35 days to reach the secular equilibrium between radon and its daughter [12]. After the irradiation period, the CR-39 films were removed from the container and chemically etched in 6.25 N solution of Na OH at 70 C^0 for 7 h. After the end of etching time, the detector were removed from the solution and washed several times with distilled water then by running water and left in air for drying .The tracks produced by alpha particles are counted using optical microscope [13] , typical optical photo for tracks has been depicted in fig1.(b). In this work, the activity concentration of radon emanated from the sample in (Bq/m^3) can be calculated from the following equation
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[14] .

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$$c = \frac{\rho}{KT} \quad (1)$$

Where C is radon concentration (Bq / m^3), K is the calibration factor to be = 0.17 (track cm^{-2}/ Bq m^{-3}d) [15]. ρ is track density (average tracks count per cm$^{2)}$,T is the exposure time. The areal exhalation rate and mass exhalation rates can be calculated from the relation [16, 17 and 19]

$$E_A = \frac{C_{Rn}\lambda V}{A\left[T+\left(\frac{1}{\lambda_{Rn}}\right)\left(e^{-\lambda_{Rn}T}-1\right)\right]} \quad (2)$$

$$E_M = \frac{C_{Rn}\lambda V}{M\left[T+\left(\frac{1}{\lambda_{Rn}}\right)\left(e^{-\lambda_{Rn}T}-1\right)\right]} \quad (3)$$

Where C_{Rn} is Radon concentration, λ_{Rn} is Radon decay constant (h$^{-1)}$, A is sample surface area (m^2), T is Exposure time (h), V is Volume of container (m^3), M is Mass of sample (kg)**.** To find a relation between the activity of radium and the track density, the specific activity of radium in 8 samples of mass 0.5 kg,has been investigated using high performance Ge detector, in lab of radiation protection at radiological authority Cairo Egypt, CR-39 films has been exposed to radon emanated from these samples with known activity of radium. The tracks formed on those films were determined using powerful optical microscope equipped with digital camera. For accurate measurements, 40 views for each sample are counted then average count for each sample is determined. Fig.2. depicts the variation of the track density as a function of the specific activity of radium. A calibration factor K for radium measurement in this work is determined to be 0.014(Track.cm^{-2}.day / (Bq. kg^{-1}).

$$C_{Ra} = \frac{\rho h A}{K M T_{eff}} \quad (4)$$

The radium activity concentration can be calculated using equation 4. (16) .where, ρ is track density, h is distance between lower surface of the detector and surface of sample, A is Surface area of sample, K is calibration factor, M is mass of sample in kg, T_{eff} is effective time, The relation between effective time and real time of exposure can be calculated from the following relation (17) .

$$T_{eff} = T.\frac{1}{\lambda(1-e^{-\lambda T})} \quad (5)$$

Where T is real time of exposure, λ is radon decay constant,

3 Results and Discussion

International regulations regarding natural radioactivity of materials take into account three main natural radionuclides: ^{40}K, ^{226}Ra and ^{228}Th. All of them are important due to their significance in ɤ-ray exposure; however, Ra-226 concentration is under special supervision because of α-alpha exposure. This exposure came from Rn-222 and its progeny. For Ra-226 concentrations limits are established in order to quantify and control α-particle exposure of Rn-222 and its progeny Po-218 and Po-214 [18]. Fig.3. Depicts the radon concentration as a function of sample number, regarding four groups. One can notice that the highest value of radon concentration is 7523.81 Bq/m^3.Also, it is clear that group A and C have the highest value of radon concentration and this may be attributed to these samples (phosphate fertilizer) containing amount of natural radioactive nuclides. On another hand, it is appear that the soil samples (group B) have the smallest value of radon concentration; this because the soil samples are consists of clay with minimum value of radioactive isotopes.

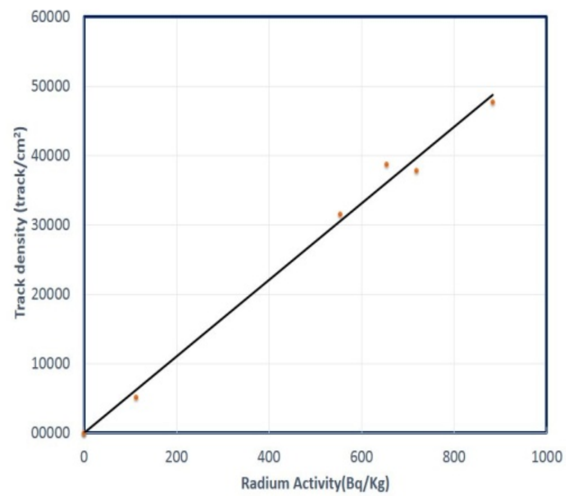

Fig.2: depicts the variation of track density as a function of radium activity.

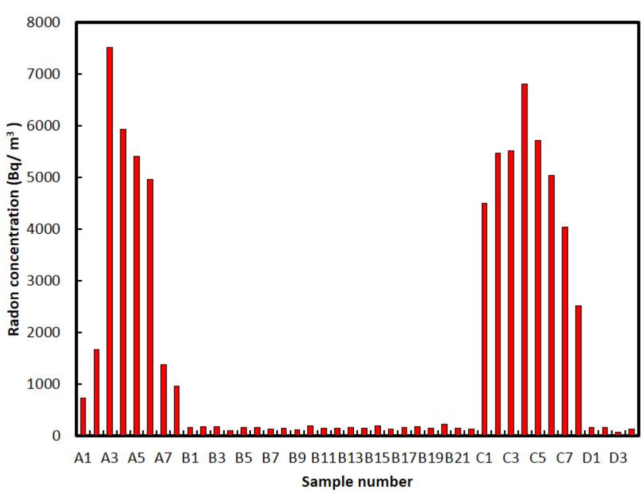

Fig.3: depicts the radon concentration emanated from the tested samples as a function of sample number.

Fig.4. depicts the radium activity as a function of sample number. One can notice that the highest value of radium activity is 969.07 Bq/kg. In addition, it is clear that group A and C have the highest value of radium activity and this may be attributed to these samples containing natural nuclides. It is appear that the soil samples (group B) has the smallest value of radium activity, this because the soil is clay sample with minimum value of radioactive isotopes. Fig.5. depicts the variation of radium activity as a function of radon concentration. A strong linear relationship has been obtained between radon and radium concentration for the measured samples.i.e. The radium isotope 226Ra content in soil is the direct source and played a key role in determining the levels of soil radon and radon exhalation from soil surface. Fig.6. depicts the mass exhalation as a function of radon concentration. Fig. 6 depicts this dependence for investigated samples, where very good linearity of the mass exhalation rate vs. radon concentration is observed. Fig. 7 depicts this dependence for investigated samples, where very good linearity of the mass exhalation rate vs. radium activity is observed.

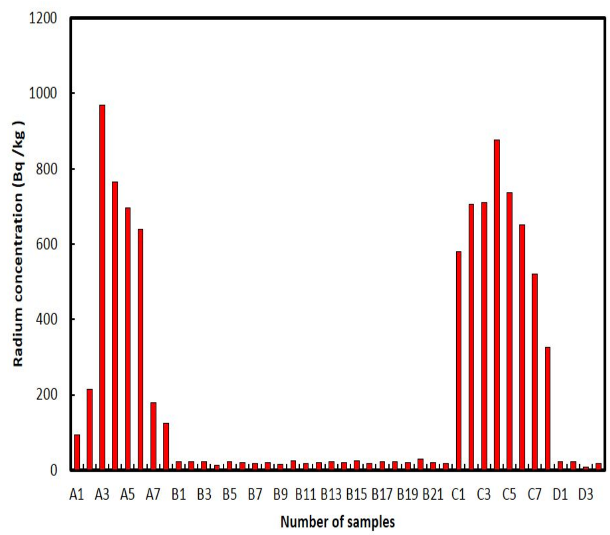

Fig.4: depicts the radium activity in the tested samples as a function of sample number.

Tables (1–4) list the ^{222}Rn activity concentration, radium content, areal and mass exhalation rate for 42 samples collected from Abo-Zaboul factory and El Khabat village. The results show that the average values of radium activity in different locations except soil and chemical fertilizers are relatively higher than normal. This survey leads to the following

1- The range of radon concentration from soil of Abo-Zaboul factory area and from soil around it is in the range from 7523.81 to 728.57 Bq/m^3 with average value of 3683.81 Bq/m^3. Also, the variation of radium in samples of Abo-Zaboul soil samples are from 969.67 to 93.84 Bq/kg with average of 474.47 Bq/kg , as shown in table 1.

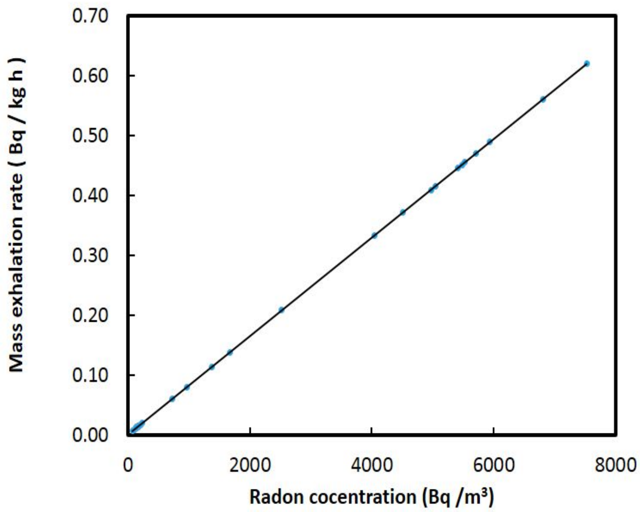

Fig.5: depicts the variation of radium concentration as a function of radon concentration emanated from the tested samples.

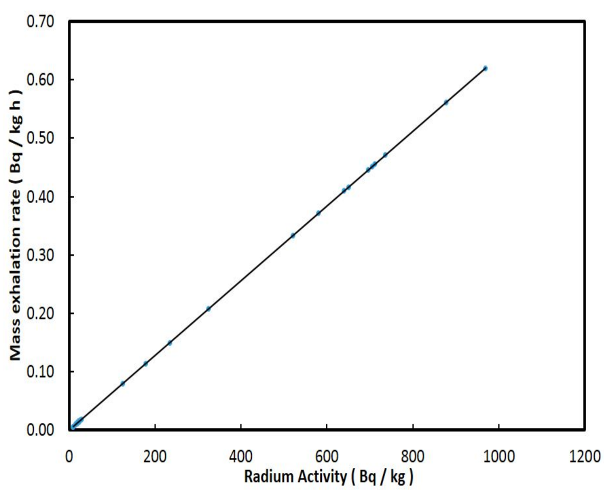

Fig.6: depicts the mass exhalation as a function of radon concentration.

2 Bq/m^3 with average value of 161.77 Bq/m^3, the radium concentration in the soil of Elkhabat village is between 29.44 to 13.49 Bq/kg with average value of 20.84 Bq/kg, as shown in table 2. The variation of radon concentration in soil samples may be attributed to the use of different kind of fertilizers used by farmers which in the recommended value declared by ICRP[22, 23].

3 The radon concentration in phosphate fertilizers are in the range of 6809.52 Bq/m3 to 2523.81 Bq/m3 with average of 4898.10 Bq/m3, the variation in radon concentration may be due to

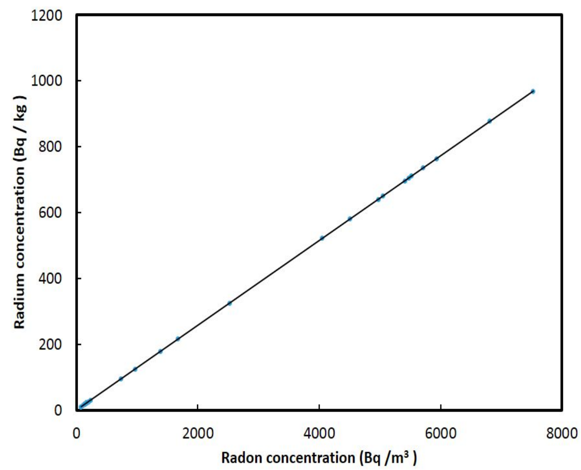

Fig.7: depicts the mass exhalation as a function of radium activity.

due to the variation in the phosphate content in each fertilizer. Also, in phosphate fertilizers the radium concentration vary from 877.07 to 325.07 Bq/kg with average value of 630.87 Bq/kg, as shown in table 3.

4 In chemical fertilizers, the radon concentration in the range from 171.43 to 71.43 Bq/m3with average value of 130 Bq/m3. And the radium concentration for chemical fertilizers varies from (22.68 to 9.20) Bq / kg with average value of 16.87 Bq / kg , as shown in table (4),which in good agreement with the value from Egypt and other countries for soil samples [25, 26, 27 ,28]

5 The obtained value of radium content in this survey is to be less than the permitted value of 370 Bq/kg reported by organization for economic and corporation and development (OECD), 1979 [26]and near the main global value of 30 Bq /kg[27]for soil and chemical fertilizers but greater than that limit for phosphate fertilizers and waste product and soil of Abo-Zaboul phosphate factory .

6 The areal exhalation rate for soil samples from Abo-Zaboul factory is in the range from 17.51 to 1.70 (Bqm-2h-1) with average value of 8.58(Bq m-2 h-1)And for soil from Elkabate village is in the range from 0.53 to 0.24(Bq m-2h-1) with average value of (0.38 Bq m-2 h-1) and for phosphate fertilizers is in the range from 15.85 to 5.87 (Bqm-2h-1) with average value of 11.40 Bqm-2h-1,and for chemical fertilizers it varies from 0.40 to 0.17 (Bqm-2h-1) with average of 0.30 Bqm-2h-1. [29, 30 ,31, 32, 33]

7 The mass exhalation rate for soil samples from Abo-Zaboul factory is in the range from 0.62 to 0.06 Bq /kg h with average value of 0.30 Bq /kg h, and for sample from Elkabat village is in the range from 0.02 to 0.01 Bq /kg h with average value of 0.01 Bq /kg h. While in phosphate fertilizers is in the range from 0.56Bq /kg h to 0.21Bq /kg h with average value of 0.40 Bq /kg h. and for chemical fertilizers it was found to be same for all for types of 0.01 Bq/kg h.[19, 20,and 21].

Table (1) ^{222}Rn activity concentration, radium content, areal and mass exhalation rate for waste and soil near Abo- Zaboulfactory (group A).

sample number	radon concentration (Bq/m^3)	radium content (Bq/kg)	areal exhalation rate ($Bq/m^2.h$)	mass exhalation rate (Bq/kg. h)
A_1	728.57±13.5	93.84±4.84	1.70±0.13	0.06±0.003
A_2	1671.43±20.44	215.28±7.34	3.89±0.20	0.14±0.02
A_3	7523.81±43.37	969.07±15.56	17.51±0.42	0.62±0.04
A_4	5933.33±38.51	764.21±13.82	13.81±0.37	0.49±0.02
A_5	5409.52±36.77	696.75±13.20	12.59±0.35	0.45±0.02
A_6	4971.43±35.25	640.32±12.56	11.57±0.34	0.41±0.016
A_7	1380.95±18.58	177.87±6.67	3.21±0.19	0.11±0.012
A_8	966.67±15.55	124.51±5.58	2.25±0.08	0.08±0.006
max	7523.81±43.37	969.07±15.56	17.51±0.42	0.62±0.04
min	728.57±13.5	93.84±4.84	1.70±0.13	0.06±0.003
average	3683.81±27.75	474.47±9.96	8.58±0.43	0.30±0.009

Table (2) show ^{222}Rn activity concentration, radium content, areal and mass exhalation rate for soil sample (1-22) from El Khabat village (group B) .

sample number	radon concentration (Bq/m^3)	radium content (Bq/kg)	areal exhalation rate $Bq/m^2.h$)	mass exhalation rate (Bq/kg. h)
B_1	166.67±6.46	21.47±2.23	0.39±0.06	0.01±0.001
B_2	180.956.73	23.31±2.41	0.42±0.06	0.01±0.001
B_3	182.38±6.75	23.49±2.42	0.42±0.05	0.02±0.001
B_4	104.76±5.12	13.49±1.84	0.24±0.04	0.01±0.001
B_5	171.43±6.55	22.08±2.35	0.40±0.06	0.01±0.001
B_6	157.14±6.27	20.24±2.35	0.37±0.05	0.01±0.001
B_7	138.10±5.88	17.79±2.11	0.32±0.05	0.01±0.001
B_8	152.38±6.17	19.63±2.22	0.35±0.06	0.01±0.001
B_9	123.81±5.56	15.95±2.00	0.29±0.04	0.01±0.001
B_{10}	200.00±7.07	25.76±2.14	0.47±0.07	0.02±0.001
B_{11}	142.86±5.98	18.40±2.54	0.33±0.05	0.01±0.001
B_{12}	152.38±6.17	19.63±2.22	0.35±0.06	0.01±0.001
B_{13}	171.43±6.55	22.08±2.35	0.40±0.06	0.01±0.001
B_{14}	152.386.17	19.63±2.22	0.35±0.06	0.01±0.001
B_{15}	200.00±7.07	25.76±2.54	0.47±0.07	0.02±0.001
B_{16}	138.10±5.88	17.79±2.11	0.32±0.04	0.01±0.001
B_{17}	166.67±6.46	21.47±2.32	0.39±0.07	0.01±0.001
B_{18}	180.95±6.73	23.31±2.41	0.42±0.06	0.01±0.001
B_{19}	152.38±6.17	19.63±2.22	0.35±0.05	0.01±0.001
B_{20}	228.57±7.56	29.44±2.71	0.53±0.07	0.02±0.001
B_{21}	152.38±6.17	19.63±2.22	0.35±0.06	0.01±0.001
B_{22}	133.33±5.77	17.17±2.07	0.31±0.04	0.01±0.001
max	228.57±7.56	29.44±2.71	0.53±0.07	0.02±0.001
min	104.76±5.12	13.49±1.84	0.24±0.04	0.01±0.001
average	161.77±6.33	20.84±2.27	0.38±0.06	0.01±001

Table (3) show ^{222}Rn activity concentration, radium content, areal and mass exhalation rate for phosphate fertilizers samples (group C).

sample number	radon concentration (Bq/m^3)	radium content (Bq/kg)	areal exhalation rate ($Bq/m^2.h$)	mass exhalation rate (Bq/kg. h)
C_1	4504.76±33.56	580.21±12.04	10.49±0.32	0.37±0.01
C_2	5476.19±37.00	705.33±13.28	12.75±0.36	0.45±0.01
C_3	5523.81±37.16	711.47±13.34	12.86±0.36	0.46±0.02
C_4	6809.52±41.26	877.07±14.81	15.85±0.40	0.56±0.03
C_5	5714.29±37.80	736.00±13.56	13.30±0.36	0.47±.02
C_6	5047.62±35.52	650.13±12.75	11.75±0.34	0.42±0.01
C_7	4047.62±31.81	521.33±11.42	9.42±0.31	0.33±0.01
C_8	2523.81±25.12	325.07±9.01	5.87±0.24	0.21±0.01
max	6809.52±41.26	877.07±14.81	15.85±0.40	0.56±0.01
min	2523.81±25.12	325.07±9.01	5.87±0.24	0.21±0.01
average	4898.10±34.90	630.87±12.53	11.40±0.34	0.40±0.01

Table (4) show ^{222}Rn activity concentration, radium content, areal and massexhalation rate for sample(1-4) group D).

sample number	Radon concentration (Bq/m^3)	Radium content (Bq/kg)	Areal exhalation rate ($Bq/m^2.h$)	Mass exhalation rate (Bq/kg. h)
D_1	166.67±6.46	21.47±2.32	0.39±0.06	0.01±0.001
D_2	171.43±6.55	22.08±2.35	0.40±0.06	0.01±0.001
D_3	71.43±4.23	9.20±1.52	0.17±0.04	0.01±0.001
D_4	133.33±5.77	17.17±2.06	0.31±0.06	0.01±0.001
max	171.43±6.55	22.08±2.35	0.40±0.06	0.01±0.001
min	71.43±4.23	9.20±1.52	0.17±0.04	0.01±0.001
average	130.95±5.75	16.87±2.06	0.30±0.06	0.01±0.001

4 Conclusions

Fertilizer and natural soil samples collected from 30 points were analyzed for natural radionuclide's using SSNTD detector. A strong positive correlation was found between radium concentration and track density registered by CR-39 Nuclear track detectiontechnique .Samples of the highest radiological hazard among tested was phosphate fertilizer, with Ra-226 concentration reaching 969.07 Bq/kg. This survey clear that, The value of the average radium concentration is less than the permitted value of 370 Bq/m^3 by (OECD) 1979 and it is less than average global value 30 Bq/kg (UNASCEAR 1993) for soil sample . The values of radon in soil samples show little variation in value, which may be due to the different types of fertilizers. The survey show strong liner relation between radium content and radon concentration in the samples. It also revel strong linear correlation between radon concentration and areal and mass exhalation rates in all samples (R^2 =1). With the data to hand, it is easy to ascribe with degree of certainty the reasons for elevated radon levels in fertilizer.

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