Determination the concentrations of radon gas and exhalation rate in some phosphate fertilizer using CR-39 track detector

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Abstract

The aim of this research is to measure the concentrations of radon gas for nine kinds of imported fertilizers samples that are widely used due to what they can cause of an increment in natural radioactive level of soil, and that will affect the increase in the environmental contamination level with radiation.

The results of the research show, the concentrations of radon gas C_{Rn} ranges from 161.9-251.98 Bqm^{-3} with the average value 208.01±32.8 Bqm^{-3}, the surface exhalation rates for radon gas Ex ranges from 0.249-0.388 $Bqm^{-2}h^{-1}$ with the average value 0.320±0.06$Bqm^{-2}h^{-1}$, the mass exhalation rates for radon gas E_m ranges from 0.183-0.285 $Bqkg^{-1}h^{-1}$ with the average value 0.235±0.04$Bqkg^{-1}h^{-1}$, the concentrations of radium gas C_{Ra} ranges from 0.365-0.57 $Bqkg^{-1}$ with the average value 0.47±0.07 $Bqkg^{-1}$ and alpha index Iα ranges from 0.0018-0.0029 with the average value 0.0023±0.0004.

From these results, the highest existence of a significant increase in the concentrations of radon gas in mono-ammonium phosphate $Jordan_1$ sample, but the lowest concentration of radon gas in ammonium phosphate Iran sample.

Key words

Radon, exhalation rate, phosphate, fertilizer, soil.

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قياس تراكيز ومعدل الانبعاث لغاز الرادون المنبعث من بعض الأسمدة الفوسفاتية باستخدام كاشف الأثر النووي CR-39

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الخلاصة

تم من خلال هذا البحث قياس تراكيز غاز الرادون C_R المنبعثمن تسع عينات من الأسمدة المستوردة المستخدمة على نطاق واسع مع التربة مما قد يسبب حدوث زيادة في مستوى النشاط الإشعاعي الطبيعي للتربة وذلك بسبب زيادة انبعاث غاز الرادون والتي من شأنها أن تعمل على زيادة نسبة التلوث البيئي ومن خلال ما سبق تم حساب كل منتراكيز غاز الرادون المنبعث وقد أظهرت النتائج أن تراكيز غاز الرادون المنبعث تراوحت بين (251.98-161.9)$Bq\ m^{-3}$ وبمعدل قدره (32.8±208.01)Bqm^{-3}. امامعدل الانبعاث السطحي Ex(s) فقد أظهرت النتائج أنتراكيز معدل الانبعاث السطحي لغاز الرادون المنبعث تراوحت قيمته بين (0.244-0.388)$Bqm^{-2}h^{-1}$ وبمعدل قدره (0.06±0.320)$Bqm^{-2}h^{-1}$. أماتراكيز معدل الانبعاث الكتليE_m تراوحت قيمته بين (0.285-0.183) $Bqkg^{-1}h^{-1}$ وبمعدل قدره (0.04±0.235)$Bqkg^{-1}h^{-1}$.
أما تراكيزغاز الراديوم الفعالC_{Ra}فقد تراوحت قيمته بين (0.57- 0.365)$Bqkg^{-1}$ وبمعدل قدره (0.47 ± 0.07)$Bqkg^{-1}$. وأخيرا تم حساب مؤثر ألفا Iα الذي تراوحت قيمته بين (0.0018 - 0.0029) وبمعدل قدره (0.0004±0.0023).
مما سبق تبين لنا ان اعلى زيادة في تركيز غاز الرادون المنبعثمن النماذج كانت واضحة في نموذج سماد ثنائي فوسفات الامونيوم المنشأ / الأردن-1 بينما كان اقل تركيز لغاز الرادون المنبعث من النماذج كانت واضحة في نموذج سماد فوسفات الامونيوم المنشأ / إيران.

Introduction

It is well known that traces of radionuclides are found in air, soil, water and human bodies, we inhale and ingest radionuclides every day of our lives and radioactive materials have been ubiquitous on earth since its creation. The presence of natural radioactivity in soil results in internal and external exposure to humans. Radioactive nuclides which can be found in nature are generally categorized in two distinct families, namely of arising from either "cosmogenic" or "terrestrial" origin. The most commonly encountered radionuclides that irradiate the human body through external exposure (primarily by gamma radiation) are ^{238}U, ^{235}U and ^{232}Th and their subsequent radioactive decay products and ^{40}K [1].

Nuclei can undergo a variety of processes which result in the emission of radiation. The most common forms of nuclear radiation are alpha, beta particles (negative and positive) and gamma-rays, our bodies contain radioactive materials such as ^{14}C and ^{40}K [2].

Although it is generally agreed that fertilizers come in two physical forms liquid and solid. The term liquid fertilizer applies to anhydrous ammonia, aqua ammonia, N solutions and liquid mixed fertilizers. Liquid N-P-K fertilizers are also known as fluid fertilizer they include true solutions which require no agitation and suspensions or slurry type mixtures of N, P and K, which require constant stirring to keep the solids suspended in the solution [3].

Collection of the samples

Nine samples of fertilizer were collected from many national marked from Baghdad city as shown in Table 1.

Table 1: Symbols of fertilizer samples.

Symbols	Fertilizer samples
S1	NPK(18,18,18) German
S2	NPK U.K.
S3	Ammonium phosphate(MAP)Jordan1-Mono
S4	Ammonium phosphate(MAP)Jordan2-Mono
S5	Soul Cyprus1Green
S6	Soul Cyprus2Green
S7	NPK (20,20,20)
S8	Ammonium phosphate(MAP)Mono-Iran
S9	Super NPK(27,27,0)

Experimental procedure

The determination of the radon gas concentrations in fertilizer samples were performed as follows, all samples were kept for one month before measurements in order to achieve the secular equilibrium for ^{222}Rn and its daughters with their respective progenies, and were performed using the nuclear track detector CR-39 of thickness of 500μm. The weight of the sample was about 5g and the detectors area were about 1×1cm^{2}, thin filter was used in order to prevent thoron gas from reaching the detector, however, leaving the radon gas to reach the detector, as shown in Fig.1.

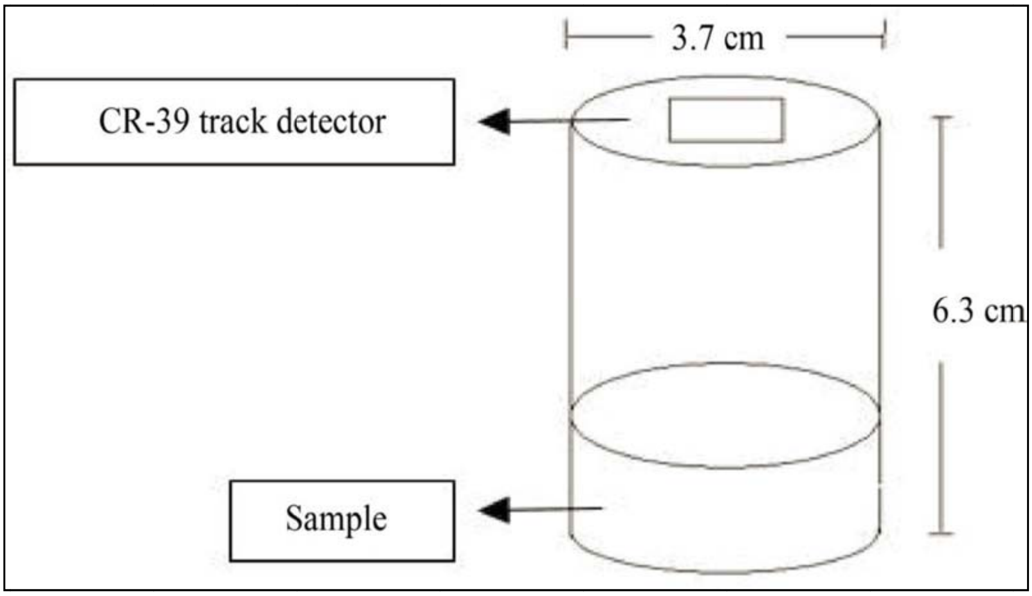

Fig.1: Sealed-cup (can) technique used for fertilizer sample.

Chemical etching process

The damaged track can be revealed through chemical etching of the material in the detector. The optimized etching conditions for CR-39 detectors were found to be 6.25 normality of sodium hydroxide NaOH solution at 60°C temperature for 6 hours [4]. Therefore, to prepare one normal solution, 1g of NaOH is dissolved in one liter of distilled water one molecular weight of NaOH can be obtained as follows:

The atomic weights of Na = 23 g/mol, O= 16 g/mol and H= 1g/mol. thus, the molecular weight of NaOH = 23 + 16 + 1 = 40 g/mol. therefore, 40 g of NaOH dissolved in 1 liter of distilled water gives 1 N solution of NaOH hence, NaOH 6.25×40 dissolved in 1 liter of distilled water gives 6.25 N solution of NaOH [5].

The etching compartment has a volume of about one liter which contains 250 g of NaOH solution with 6.25N. The condenser tube is a closed assembly except for a small vent at the top, which prevents any change of etchant normality (concentration) during the experiment as a result of evaporation. The CR-39 detectors were placed in the etching solution as shown in Fig. 2.

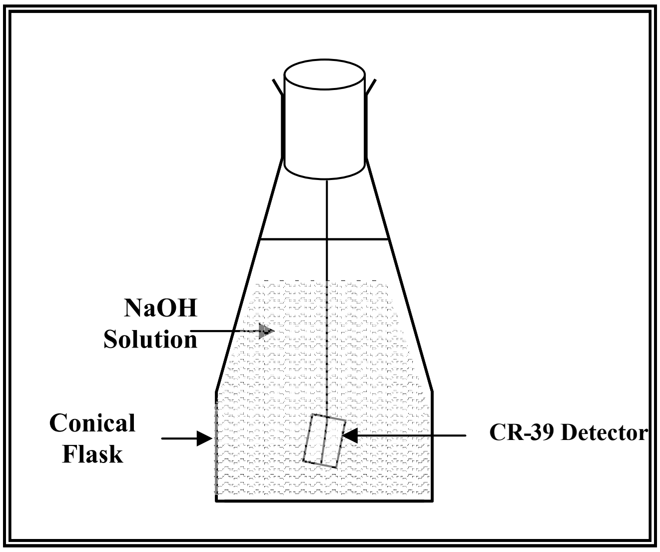

Fig.2: Chemical etching process for CR-39 track detector.

Measurements of tracks density

The tracks density was measured using this relation:

Tracks density (ρ) =

$$\frac{\text{Average number of total pits(track)}}{\text{Area of field view}} \quad (1)$$

an example of the photograph of observed tracks in the samples is shown in Fig.3.

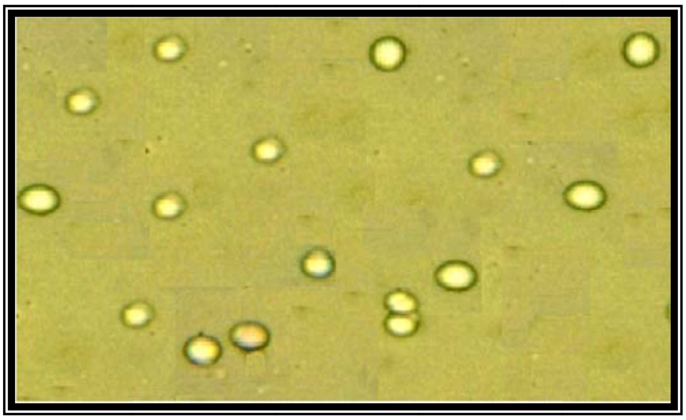

Fig.3: Photograph of tracks density.

After the irradiation time 30 days, the tracks density was recorded using an optical microscope with a magnification of 400 x. The density of the alpha particles tracks (ρ) in the samples were calculated according to relation 1.

The radon gas concentration in fertilizer samples were obtained by the comparison between track densities registered on the detectors of the sample and that of the standard samples, using the relation [6]:

$$C_X = \rho_X * (C_S / \rho_S) \quad (2)$$

where:

C_X: is the radon gas concentration in the unknown sample.

C_S: is the radon gas concentration in the standard sample.

ρ_X: is the track density of the unknown sample (track/mm^2).

ρ_S: is the track density of the standard sample (track/mm^2).
The unknown concentration was obtained using the following relation: standard uranium weight (known) × concentration (known) = weight of the sample (known) × unknown concentration. Fig.4 represents the obtained calibration curve for standard samples.

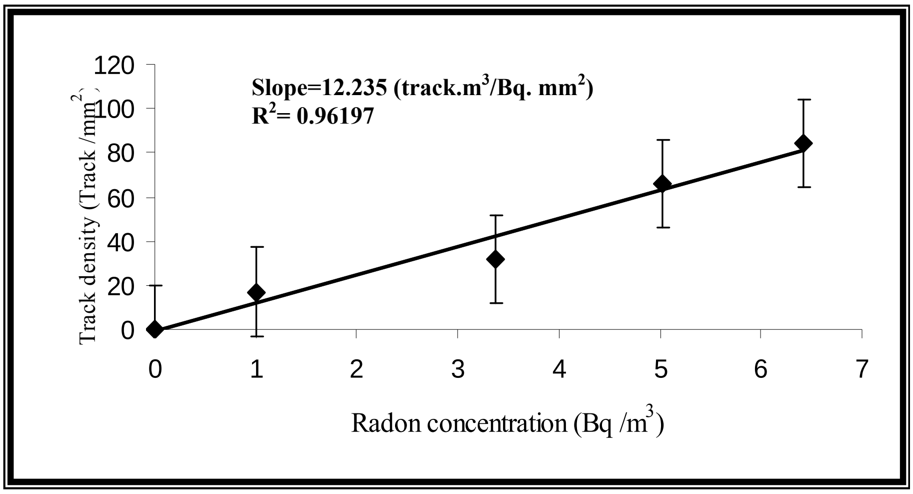

Fig. 4: The relation between radon gas concentration and track density in standard samples [7].

The effective radium content C_{Ra} in samples were calculated as; [8]:

$$C_{Ra} = \frac{\rho * h * A}{k * M * Te} \quad (3)$$

where h: the distance from the sample to CR-39 detector.
A: the area of the cup.
K: the calibration factor which is equal to (3.9816 tracks.mm^{-2}d^{-1}/Bq.m^{-3})
Te: the effective time calculated as Te = [T-λ_{Rn}^{-1}(1- e$^{-\lambda RnT}$)]
T: exposure time.
The radon exhalation rate in terms of area E_x in (Bq.m^{-2}.h^{-1}) was calculated as follows [8, 9]:

$$E_x = \frac{CV\lambda}{A[T + \lambda^{-1}(e^{-\lambda T} - 1)]} \quad (4)$$

where C: the integrated radon exposure measured by the CR-39 SSNTD (Bq.m^{-3}.h).
V: the effective volume of the container (m^3).
λ: the decay constant of radon (h^{-1}).
T: the exposure time (h), and A is the area covered by the container (m^2).

The radon exhalation rate in terms of mass E_m in (Bq kg^{-1}h^{-1}) was calculated as:

$$E_m = \frac{CV\lambda}{M[T + \lambda^{-1}(e^{-\lambda T} - 1)]} \quad (5)$$

where M is the mass of the sample (kg), the alpha index was calculated as [10].

$$I\alpha = C_{Ra} / 200 \quad (6)$$

Finally the concentrations of radon gas in fertilizer samples are shown in Table 2.

Results
Fig. 5 shows the lowest of radon concentration was found in (S8) is equal to (161.9Bqm^{-3}), the highest of radon concentration was found in (S3) is equal to (251.98Bqm^{-3}) and the average value is equal to (208.01± 32.8Bqm^{-3}).
Fig. 6 shows the lowest of surface exhalation rate was found in (S8) is equal to (0.249Bq m^{-2}h^{-1}), the highest

of surface exhalation rate was found in (S3) is equal to (0.388Bq $m^{-2}h^{-1}$) and the average value is equal to (0.320±0.06 Bq m^{-2} h^{-1}).

Fig. 7 shows the lowest of mass exhalation rate was found in (S8) is equal to (0.183Bq kg^{-1} h^{-1}), the highest mass exhalation rate was found in (S3) and equal to (0.285Bq kg^{-1} h^{-1}) and the average value is equal to (0.235 ± 0.04 Bq kg^{-1} h^{-1}).

Fig. 8 shows: the lowest of radium concentration was found in (S8) is equal to (0.365 Bq kg^{-1}), the highest of radon concentration was found in (S3) is equal to (0.57 Bqkg^{-1}) and the average value is equal to (0.47 ± 0.07 Bq kg^{-1}).

Fig. 9 shows the lowest of alpha index was found in (S8) and equal to (0.0018), the highest of alpha index was found in (S3) equal to (0.0029) and the average value is equal to (0.0023 ±4x10^{-4}).

These results shows that, the average values of radon gas concentration C_{Rn} in fertilize samples (208.01± 32.8 Bqm^{-3}) greater than the value of the global limit (200 Bqm^{-3}), and the averages radiological hazard index (C_{Ra}, E_x, E_m and Iα) in fertilize samples are, (0.47±0.07Bq kg^{-1},0.320±0.06Bq m^{-2} h^{-1}, 0.235±0.04 Bq kg^{-1} h^{-1} and 0.0023 ±4x10^{-4}) respectively.

Table 2: Radon gas concentration, surface exhalation rate, mass exhalation rate, radium concentration and alpha index for fertilizer samples.

Symbol	C_{Rn} Bqm^{-3}	E_x Bq$m^{-2}h^{-1}$	E_m Bqkg^{-1} h^{-1}	C_{Ra} Bqkg^{-1}	I_α
S1	179.89	0.277	0.203	0.4	0.0020
S2	180.78	0.278	0.204	0.41	0.0021
S3	251.98	0.388	0.285	0.57	0.0029
S4	174.35	0.268	0.197	0.393	0.0019
S5	207.55	0.319	0.234	0.468	0.0023
S6	234.13	0.36	0.264	0.528	0.0026
S7	249.07	0.383	0.281	0.562	0.0028
S8	161.9	0.249	0.183	0.365	0.0018
S9	232.47	0.360	0.326	0.525	0.0026
average	208.01±32.8	0.320±0.06	0.235±0.04	0.47±0.07	0.0023 ±4×10^{-4}

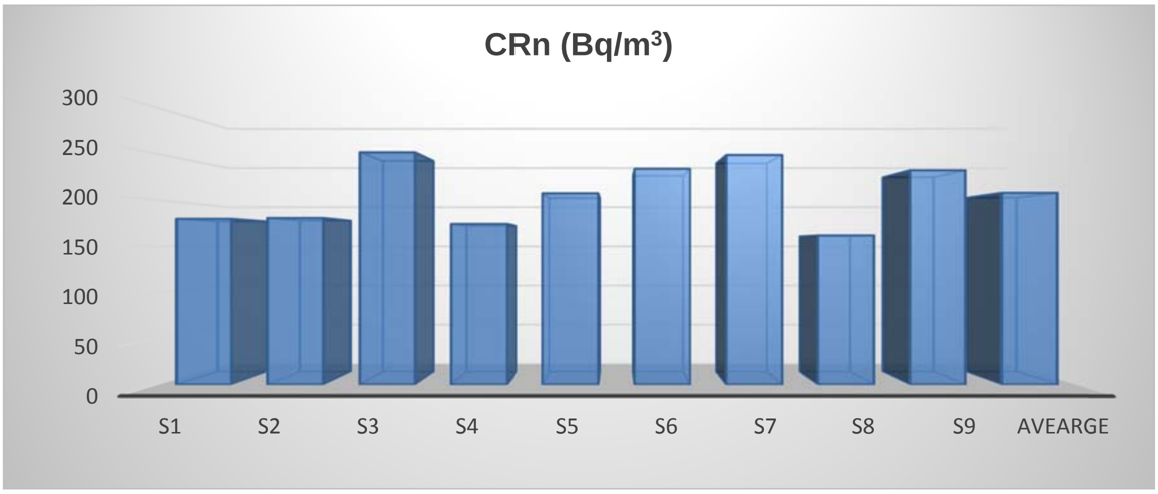

Fig.5: Concentrations of radon gas for fertilizer samples.

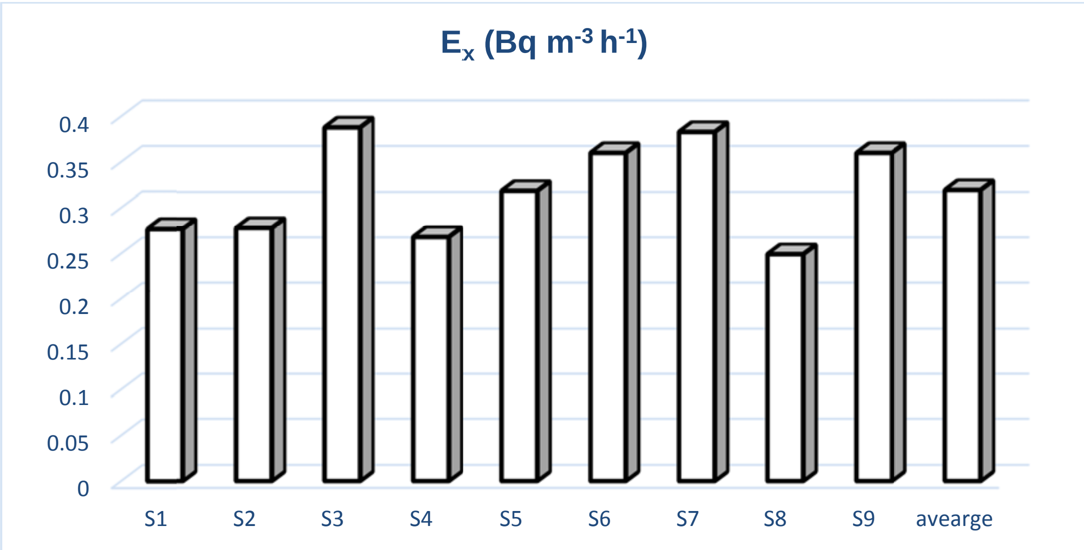

Fig.6: Surface exhalation rate for fertilizer samples.

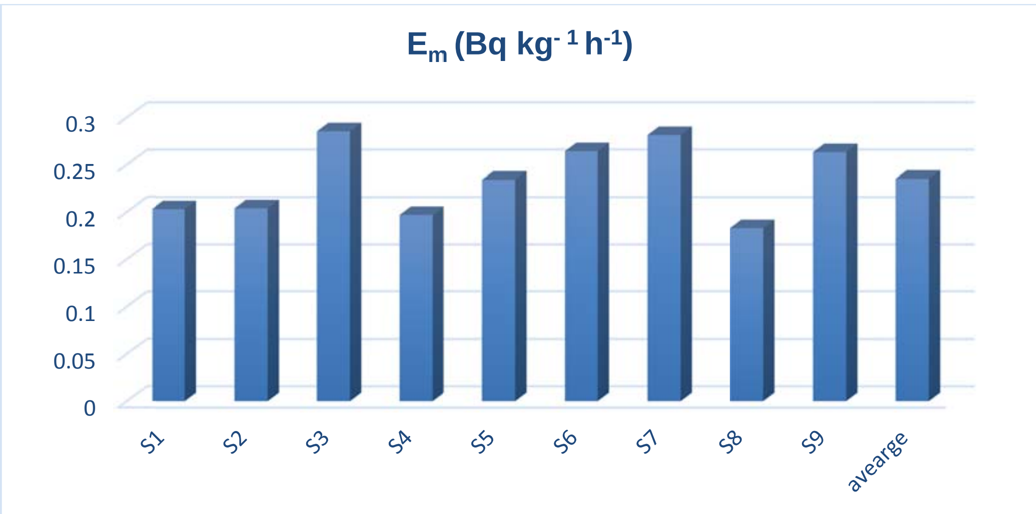

Fig.7: Mass exhalation rate for fertilizer samples.

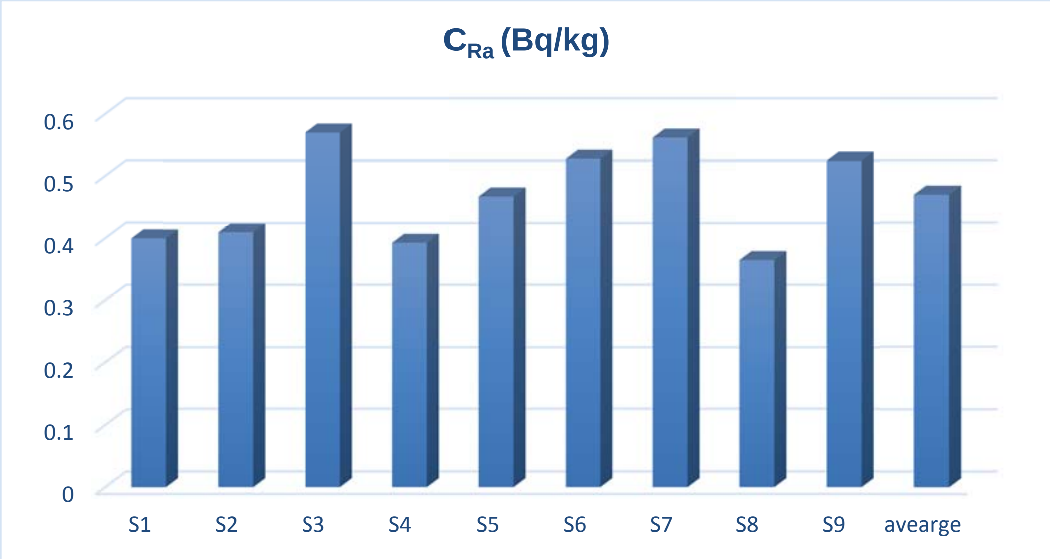

Fig.8: Radium gas concentration for fertilizer samples.

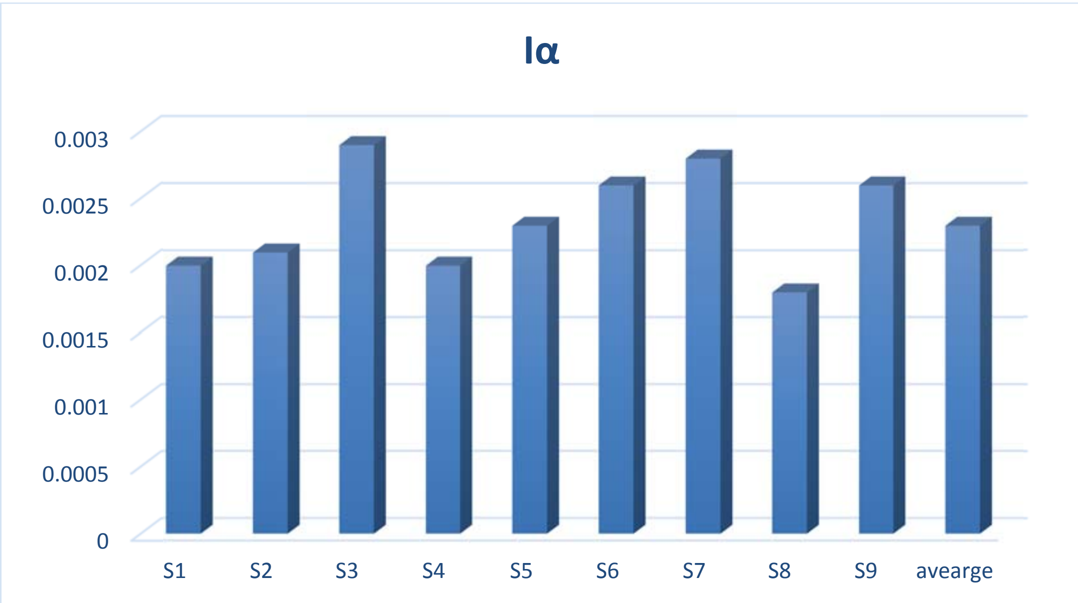

Fig.9: Alpha index for fertilizer samples.

Conclusions

The maximum value of radon concentration in fertilizer samples was found in Ammonium phosphate (MAP) Mono-Jordan which made in Jordan1 and gives a large value of (C_{Ra}, E_x, E_m and Iα), the highest radon concentration in fertilizer samples was found in Ammonium phosphate (MAP) Mono-Jordan1equal to (251.98Bqm^{-3}), the reason of the highest concentration may be that the fertilizer containing high concentrations of uranium. The average of radon gas concentration was greater than the global limit (200Bqm^{-3}).

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