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Natural Radioactivity and Radon Exhalation in Phosphate Fertilizers

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Abstract ^{238}U , ^{232}Th , ^{226}Ra , ^{222}Rn and ^{40}K activity concentrations in fertilizer samples collected from a local market were determined. Radon activity concentrations ranged from 0.04–252 Bq kg⁻¹ and were comparable with low activity concentrations ranges reported previously. Radon exhalation rates and effective ^{226}Ra concentration were determined using CR-39 solid-state nuclear track detectors. Effective ^{226}Ra concentrations ranged from 4.27–33.93 Bq kg⁻¹. Two different ^{222}Rn exhalation rates were determined; area dependent exhalation rates (0.044–0.351 Bq m⁻² h⁻¹) and mass dependent exhalation rates (0.023–0.180) Bq kg⁻¹ h⁻¹. Even without secular equilibrium, strong correlations were generally apparent among uranium concentrations, effective ^{226}Ra concentrations and ^{222}Rn exhalation rates in samples with high concentrations of uranium. ^{232}Th and ^{40}K concentrations ranged from 0–44.6 and 0.08–127 µg/g, respectively.

Keywords Natural radioactivity · ^{238}U · ^{232}Th · ^{226}Ra · ^{222}Rn · ^{40}K · Radon exhalation · Fertilizers

الخلاصة

تم قياس تركيز كل من ^{238}U ، ^{232}Th ، ^{226}Ra و ^{40}K في عينات من السماد الفوسفاتي التي جمعت من السوق المحلي، حيث وجد أن إشعاعية الرادون تقع في المدى 0.04–252 Bq kg⁻¹ وهو ما يقارن بالتركيز المنخفضة المذكورة في المراجع. وقد تم أيضا قياس معدل انبعاث الرادون وتركيز الراديوم الفعال باستخدام الكواشف النووية الصلبة من نوع CR-39. وتقع تراكيز الراديوم الفعالة في المدى 4.27–33.93 Bq kg⁻¹ كما تم ايجاد نوعين من مقدار انبعاث الرادون. والانبعاث المعتمد على المساحة E_A (0.044–0.351) Bq m⁻² h⁻¹ والانبعاث المعتمد على الكتلة E_M (0.023–0.180) Bq kg⁻¹ h⁻¹. مع غياب الاتزان فإنه لا تزال هناك علاقة واضحة بين تركيز اليورانيوم وتركيز الراديوم الفعال ومعدل انبعاث الرادون للعينات التي تحتوي على تراكيز مرتفعة نسبيا من اليورانيوم. وتقع تراكيز كل من ^{232}Th و ^{40}K في المدى (0–44.6µg/g) و (0–127µg/g) على الترتيب.

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1 Introduction

Natural radioactivity mainly results from the natural decay series and singly occurring radioisotopes like ^{40}K and ^{14}C . More than 70% of radiation exposure to the general population is from natural sources, with radon as the major source of exposure. Other radiation sources include cosmic rays and isotopes within the human body [1]. Three natural decay series originate with isotopes of uranium and thorium, resulting in isotopes of lead [2]. In an ideal situation, species in the natural decay chain are in equilibrium when the equilibrium conditions are met. Often in nature these equilibrium conditions are not met, typically resulting from leaching of one or more members out of the chain. For example, uranium can oxidize from U (IV) to U (VI) under oxidizing conditions. This oxidation produces water soluble uranyl complexes, resulting in the removal of uranium from the decay chain and a disruption of the natural equilibrium. Natural uranium is present as three isotopes: ^{234}U (0.00059–0.005%), ^{235}U (0.7198–0.7202%) and ^{238}U (99.2739–99.2752%). Uranium is an important raw material for nuclear energy production and other industrial applications. Along with its radioactive properties, uranium is also chemically toxic. Precautions should be taken against the inhalation of uranium dust, which has a threshold limit in air of 0.20 mg m^{-3} . The maximum daily uranium intake set by the World Health Organization (WHO) is $0.6\text{ }\mu\text{g kg}^{-1}$ of body weight. The WHO guidelines for drinking water are $30\text{ }\mu\text{g L}^{-1}$. The maximum uranium concentration in drinking water should not exceed $20\text{ }\mu\text{g L}^{-1}$. The earth's crust contains $3\text{--}4\text{ mg kg}^{-1}\text{ U}$, making it approximately as abundant as arsenic or boron [3,4].

Radon is a chemically inert noble gas. Three isotopes of radon form the natural decay series: ^{222}Rn ($t_{1/2} = 3.82$ days), ^{220}Rn (thoron, $t_{1/2} = 55.6$ s) and ^{219}Rn (actinon, $t_{1/2} = 3.96$ s). All three Rn isotopes are alpha-particle emitters. Radon can escape from soil, rocks and water that contain its immediate parent nuclide, radium. The short half-lives of ^{220}Rn and ^{219}Rn mean that they decay before they have a chance to be released into the atmosphere, generally posing little risk to the population. Conversely, with a half-life of 3.8 days, ^{222}Rn is likely to escape its immediate surroundings and enter the atmosphere. When released into the atmosphere and inhaled, the relatively long half-life of ^{222}Rn means it is exhaled from the lungs before it decays. Since on average, inhaled air remains in the body for less than a minute, the decay of ^{222}Rn is negligible. Short-lived daughter species of radon, however, have a range of half-lives from $1.6 \times 10^{-4}\text{ s}$ –26.8 min. When these daughter species are produced, they are electrically charged and adsorb to aerosols present in the atmosphere, which enter the body through inhalation. Being short-lived, these daughter species are likely to decay before removal from the lung, a process taking up to 30 min. This pathway is likely responsible for the strong association of radon exposure with lung cancer [5,6]. It has been suggested that ionizing radiation from the short-lived decay products of radon affect bronchial cells, initiating cancers of the respiratory tract, especially lung cancer. Evidence linking exposure to high levels of radon and an increased risk of lung cancer is mounting [7]. Recent epidemiological studies have consistently shown statistically significant increases in lung cancer risk resulting from exposure to radon in residential buildings at moderate exposures, and strong synergistic effects with cigarette smoking [8]. Sufficient data has shown that radon and its decay products are the principal causes of lung and other related cancers [9].

Phosphate deposits generally display enhanced radionuclide concentrations compared to natural levels. Uranium in phosphate deposits occurs as complexes with calcium phosphate. The mining and processing of phosphate ores redistribute natural radionuclides in the environment, making them available in ore products such as phosphogypsum (PG) and phosphoric acid. Phosphoric acid is a starting material for triple superphosphate (TSP), monoammonium phosphate (MAP), diammonium phosphate (DAP), NPK fertilizers and dicalcium phosphate (DCP). Radionuclides from natural decay series are present in all processes in the phosphate industry and this has prompted numerous studies on the presence and effects of radionuclides. Saueia et al. [10] studied radionuclide concentrations by performing radiological characterizations on phosphate rocks, phosphogypsum and phosphate fertilizers in Brazil using alpha- and gamma-spectrometry. Their study concluded that fertilizer samples derived directly from phosphoric acid (MAP and DAP) showed low activities for ^{226}Ra , ^{228}Ra and ^{210}Pb and significant concentrations of U and Th, up to 822 and 850 Bq kg^{-1} , respectively. Fertilizers produced by mixing phosphoric acid and phosphate rock (SSP and TSP) had higher concentrations of radionuclides, for example, 1,138 Bq kg^{-1} for ^{238}U and 521 Bq kg^{-1} for ^{232}Th [11]. Al-Jundi et al. [12] investigated activity concentrations of ^{238}U , ^{226}Ra , ^{228}Ra , ^{210}Pb and ^{40}K in Jordanian phosphogypsum and fertilizers. The activity concentration of ^{238}U had mean values of $1033 \pm 22\text{ Bq kg}^{-1}$ in fertilizer samples and $22 \pm 11\text{ Bq kg}^{-1}$ in phosphogypsum samples. Tufail et al. [13] investigated the specific activity of ^{226}Ra in phosphate rocks from Jordan and Pakistan and found the specific activities to be 428 ± 11 and $799 \pm 10\text{ Bq kg}^{-1}$, respectively.



Phosphate fertilizers and soil are also active areas of research. Ghosh et al. [14] measured natural radioactivity in fertilizer and agricultural soil, finding a positive correlation between alpha-activity in soil and fertilizer samples. Naturally occurring radionuclides can enter soils and groundwater from human practices like the application of certain fertilizers for agricultural purposes. Soils receiving phosphate fertilizers accumulate U, Th and their daughter nuclides, which can eventually leach into groundwater [15]. Low concentrations of ^{226}Ra and ^{232}Th isotopes can enter the environment through discharged wastewater from the phosphate fertilizer industry [16].

2 Experimental

2.1 Sampling and Sample Preparation

Samples of 12 widely used fertilizers were collected from a local market in Jeddah, Saudi Arabia. Commercial names and phosphate and potassium concentrations in the fertilizer samples are summarized in Table 1. Phosphorous in the fertilizers was present as P_2O_5 , with concentrations ranging from 7–84%. Samples 1–9 and 12 were solids with different particle sizes. The samples were crushed into fine powders using a mortar and pestle.

2.2 Uranium, Thorium and Potassium Concentrations

Uranium and thorium concentrations in the fertilizer samples were determined using a Perkin–Elmer ICP-MS spectrometer (Sciex Elan DRCII, USA). The ICP-MS instrument was calibrated daily before measurements and operated as recommended by the manufacturer. Potassium was determined using an Analyst 800 Perkin–Elmer atomic absorption spectrometer (AAS). Analytical reagent grade chemicals were used for all preparations. Solutions were prepared using deionized water produced in the lab from a Milli-Q plus system (Millipore, Bedford, MA, USA). This system produced deionized water with a resistivity of less than $18.2 \text{ M}\Omega \text{ cm}$ at 25°C .

Fertilizer samples were digested in concentrated HNO_3 . The fertilizer digestion was performed by weighing 0.2 g of the fertilizer sample into a beaker and adding 10 mL of HNO_3 . After standing for 2 days, the solution was heated at 70°C to near dryness. Then, 10 mL of 5% HNO_3 was added and the solution was heated again to ensure no solid residue remained. The remaining solution was analytically transferred to a 25 mL polyethylene volumetric flask for analysis. The polyethylene flasks were cleaned prior to use with a detergent, soaked in 5% HNO_3 and rinsed with deionized water.

To verify the analytical procedure, the certified reference material (CRM) Soil-7 of the IAEA was used. The CRM was digested using conc. HNO_3 and conc. HF by transferring 0.10 g of the CRM to a 100 mL beaker and adding 7 mL of conc. HNO_3 and 2 mL of conc. HF. After standing for 24 h, the solution was heated at 70°C until near dryness and 5 mL of conc. HNO_3 was added. The heating and acid addition steps were repeated twice more to ensure that all solids were dissolved. The digested solution was analytically transferred to a 25 mL polyethylene volumetric flask for analysis.

Table 1 Properties of the fertilizer samples

Sample	Type	Contents		Density (Kg m^{-3})
		% P_2O_5	% K_2O	
F1	Granular	20	20	1.01×10^3
F2	Granular	0	0	1.06×10^3
F3	Granular	12	17	1.04×10^3
F4	Granular	18	5	1.11×10^3
F5	Granular	0	0	0.77×10^3
F6	Granular	18	5	1.02×10^3
F7	Granular	7	8	0.56×10^3
F8	Granular	30	15	0.93×10^3
F9	Granular	20	20	0.61×10^3
F10	Solution	8	6	1.13×10^3
F11	Solution	P (50 g/L)	K (50 g/L)	1.13×10^3
F12	Leafy	48	8	0.74×10^3



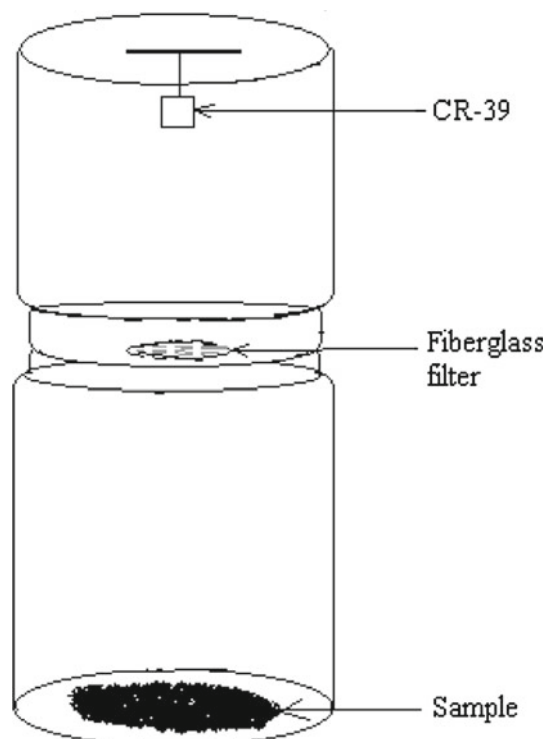


Fig. 1 Schematic representation of the radon measurement detection chamber

2.3 Radon Measurements

Solid-state nuclear track detectors, commercially known as CR-39 detectors, manufactured by Track Analysis Systems Ltd, UK, were used for the determination of radon concentrations. Detectors, 2 cm × 2 cm in size, were delivered in sealed foil containers. Radon was measured using a detection chamber assembled in our lab. The detection chamber consisted of a CR-39 detector manufactured to hang in an empty 750 mL glass container. The container was mounted upside-down on a similar container and a lid was placed between the two containers. One round hole (2.5 cm radius) was made in the middle of the lid. Glass fiber filter paper (Ahlstrom 121) was placed over the hole. This filter allowed ^{222}Rn to diffuse into the detector-containing chamber and discriminated against the shorter-lived ^{220}Rn isotope. The filter also limited the humidity in the detection chamber. This procedure measured primary alpha-particles and short-lived daughter species from radon originating inside the container. A schematic representation of the detection chamber is presented in Fig. 1. Two grams of fertilizer sample was placed in the lower glass container and the detection system was set up and completely sealed to avoid any leaks. The detectors were exposed to radon present in the fertilizer samples for three months, after which they were removed and processed.

After exposure to radon, the detectors were removed from the detection chamber and etched using a 20% KOH solution at 70°C. Etching was performed in a temperature regulated water bath where a beaker containing 20% KOH were placed in the water bath. The detectors were immersed in the solution, with both sides exposed to the etchant, for 6 h. After etching, the detectors were removed and cleaned using 0.1 M HCl at 70°C once and deionized water twice. The detectors were then wiped clean using isopropyl alcohol wipes and stored in sealed bags for track counting.

Tracks were manually counted using an optical microscope (Axiostar-Carl Zeiss Microimaging GmbH), using 50 × objectives. A 7.1 megapixel Canon PC1089 PowerShot G6 camera was attached to the microscope to capture the field of magnification as an image file. The field-of-view was also observed directly on a computer screen attached to the microscope.

2.3.1 Standard Solution

A 1 mL standard solution of ^{226}Ra with an absolute activity of 21.7288 Bq mL⁻¹ was used to determine the response function of the detection system. In the passive determination of radon, no standard reference material



is available. Each radon-containing material has its own unique characteristics. Quality assurance was achieved through the use of a standard radon source (generated from radium), a check of linearity in the response of the detection system and blind tests. For calibration purposes, four detectors were separately exposed to radon from a ^{226}Ra source for periods of 3, 5, 7 and 14 days. After each exposure the detector was removed, etched, cleaned and tracks were observed and counted.

3 Results and Discussion

3.1 ^{238}U , ^{232}Th , and ^{40}K Concentrations

A summary of ^{238}U , ^{232}Th and ^{40}K concentrations in fertilizers is presented in Table 2. Samples F2 and F5 contained no phosphorous and, as a result, their uranium concentrations were very low. Sample F10 contained 8% P_2O_5 , but the observed uranium concentration was comparable to samples F2 and F5. This result from sample F10 being a liquid. Sample F11 was also a liquid and had an observed uranium concentration of $40 \mu\text{g kg}^{-1}$. Sample F12 contained a relatively low concentration of uranium ($638 \mu\text{g kg}^{-1}$) despite containing 48% P_2O_5 . Sample F12 was a leafy fertilizer and the uranium concentration is comparable to results reported by Ashraf and Khater [17]. Table 3 summarizes the specific activities of uranium in various countries. Activities observed in this work lie in the lower range of activities reported previously.

3.2 Radon

Radon exhalation rates were calculated using Equations (1) and (2) [18, 19]:

$$E_M(\text{Bq kg}^{-1} \text{ h}^{-1}) = \frac{CV\lambda/M}{T + 1/\lambda(e^{-\lambda T} - 1)} \tag{1}$$

$$E_A(\text{Bq m}^{-2} \text{ h}^{-1}) = \frac{CV\lambda/A}{T + 1/\lambda(e^{-\lambda T} - 1)} \tag{2}$$

Table 2 ^{238}U , ^{232}Th and ^{40}K concentrations (with associated standard deviations) in fertilizer samples

Sample	Concentration ($\mu\text{g/kg}$)		
	^{238}U	^{232}Th	^{40}K
F1	31.0 ± 0.03	0.55 ± 0.67	126.87 ± 3
F2	5.0 ± 0.02	<LOD	0.09 ± 0.03
F3	11075.0 ± 0.1	8.34 ± 0.98	118.22 ± 7
F4	10128.0 ± 0.18	6.99 ± 0.26	35.62 ± 2
F5	7.0 ± 0.01	6.84 ± 0.16	0.08 ± 0.7
F6	19950.0 ± 0.05	5.76 ± 0.02	56.97 ± 2.5
F7	7965.0 ± 0.06	1.44 ± 0.31	111.14 ± 2
F8	34.0 ± 0.02	4.71 ± 0.15	120.33 ± 7
F9	197 ± 0.01	13.124 ± 0.59	80.6 ± 1
F10	3 ± 0.03	12.151 ± 0.18	37.5 ± 1
F11	40 ± 0.01	Below the detection limit	34.7 ± 2
F12	638 ± 0.04	44.563 ± 0.25	79.3 ± 0.6

LOD lower limit of detection

Table 3 Specific activity of uranium reported in various countries

Location and type	Description	U (Bq kg^{-1})	References
Germany	Commercial	2,375.4–3,418.8	[20]
Croatia	Fertilizer plants	819.0–3,186.0	[21]
Turkey fertilizer plants	NPK	272.2–345.3	[22]
	DAP	463.0–754.6	
Brazil fertilizer plants		27.5–686.9	[23]
Egypt fertilizer plants		86.9–1,490.3	[24]
KSA commercial	Granular	174.0–2,234	[17]
	Leafy	<20	
KSA commercial		0.04–252.37	This work



Table 4 Radon activity and exhalation rates of fertilizer samples

Sample	Tracks (cm ⁻²)	Activity (Bq m ⁻³)	Activity (Bq kg ⁻¹)	E _A (mBq m ⁻² h ⁻¹)	E _M (mBq kg ⁻¹ h ⁻¹)
F1	290 ± 17	519.14	785.85	61.29	31.43
F2	263 ± 16	470.80	712.68	55.58	28.50
F3	1,659 ± 41	2,969.82	4,495.59	350.60	179.79
F4	1,404 ± 37	2,513.33	3,804.59	296.71	152.16
F5	213 ± 15	381.30	577.19	45.01	23.08
F6	422 ± 21	755.43	1,143.54	89.18	45.73
F7	1,032 ± 32	1,847.41	2,796.53	218.09	111.84
F8	268 ± 16	479.75	726.23	56.64	29.05
F9	345 ± 19	617.59	934.89	72.91	37.39
F10	209 ± 14	374.14	266.35	44.17	22.65
F11	218 ± 15	390.25	590.74	46.07	23.63
F12	227 ± 15	406.36	615.13	47.97	24.60

Table 5 Uranium and effective radium concentrations

Sample	Concentration (Bq kg ⁻¹)		Effective C _{Ra} CR-39
	C _U ICP-MS	C _{Rn} CR-39	
F1	0.39	785.85	5.93
F2	0.06	712.68	5.38
F3	140.10	4,495.59	33.93
F4	128.12	3,804.59	28.71
F5	0.9	577.19	4.36
F6	252.37	1,143.54	8.63
F7	100.76	2,796.53	21.10
F8	0.43	726.23	5.48
F9	2.49	934.89	7.06
F10	0.04	266.35	4.27
F11	0.51	590.74	4.46
F12	8.07	615.13	4.64

where E_M is the mass exhalation rate, E_A is the surface exhalation rate, C is the integrated radon exposure, T is the exposure time, V is the volume of the container, M is the mass of the sample, A is the surface area of sample and λ is the decay constant of ²²²Rn. Observed radon activities (Bq m⁻³), (Bq kg⁻¹) and radon exhalation rates, E_A and E_M, are presented in Table 4.

In general, comparable radon activity concentrations and exhalation rates were observed for fertilizers analyzed by Saad [25].

3.3 Effective Radium Concentrations and Relation to Uranium Concentrations

The effective radium concentration was calculated using Equation (3) [26].

$$C_{Ra} = \frac{\rho V}{\eta M T_{eff}} \quad (3)$$

In Equation 3, ρ is the observed track density, V is the effective volume of the container (m³), T_{eff} is the effective exposure time [T_{eff} = T + 1/λ (1 - e^{-λT})] and M is the mass of the sample (kg). The detection efficiency of the CR-39 detectors was calculated from the observed tracks (cm⁻²) per hour and the activity (m⁻³) and was equal to η = 0.016 tracks cm⁻² h⁻¹/Bq m⁻³. Table 5 presents a summary of uranium and effective radium concentrations from the radon measurements. All of the effective radium activities were different than the uranium activities in the fertilizer samples, indicating the absence of a secular equilibrium. Samples F1, F2, F5 and F8–F11 had higher radium activities, while samples F3, F4, F6, F7 and F12 had lower radium than uranium activities. These results indicate different mechanisms and quantities of isotopes were removed from the general equilibrium. Except in sample F6, there were strong correlations among uranium concentrations and both exhalation rates and effective radium concentrations. Generally these correlations were clearer at high concentrations of uranium, but less clear for low concentrations.



4 Conclusions

Uranium concentrations were determined for fertilizer samples collected from a local market. Uranium activity concentrations ranged from 0.04–252 Bq kg⁻¹. This range was comparable to the lower end of ranges reported previously. Radon exhalation rates and effective ²²⁶Ra concentrations were determined using CR-39 solid state nuclear track detectors. Effective ²²⁶Ra concentrations ranged from 4.27–33.93 Bq kg⁻¹. Two different ²²²Rn exhalation rates were determined; area dependent exhalation rates (E_A, 0.044–0.351 Bq m⁻² h⁻¹) and mass dependent exhalation rates (E_M, 0.023–0.180 Bq kg⁻¹ h⁻¹). Even in the absence of a secular equilibrium, strong correlations were generally observed among uranium concentrations and effective ²²⁶Ra concentrations and ²²²Rn exhalation rates in samples with high concentrations of uranium. ²³²Th and ⁴⁰K concentrations ranged from 0–44.6 and 0.08–127 μg/g, respectively.

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