



ALPHA PARTICLES EMISSIONS IN SOME SAMPLES OF MEDICAL DRUGS (CAPSULE) DERIVED FROM MEDICAL PLANTS IN IRAQ

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Abstract

In present study, Radioactivity of ^{222}Rn , ^{226}Ra and ^{238}U were measured in 22 samples of medical drugs (capsule) derived from medical Plants in Iraq using a solid-state nuclear track detector (LR-115 Type II). Also, the annual average internal effective dose and risk of an excess cancer fatality per million person due to ingestion of ^{222}Rn , ^{226}Ra and ^{238}U were estimated. The results show that, radon concentrations were ranged from 0.51 Bq/m^3 to 60.16 Bq/m^3 with an average 16.78 Bq/m^3 , radium concentrations were ranged from $1.69 \mu\text{Bq/kg}$ to $795.51 \mu\text{Bq/kg}$ with an average $145.95 \mu\text{Bq/kg}$, while uranium concentrations were ranged from $0.02 \mu\text{Bq/kg}$ to $8.91 \mu\text{Bq/kg}$ with an average $1.64 \mu\text{Bq/kg}$. The total average values of annual average internal effective dose and risk of an excess cancer fatality per million person due to ingestion of ^{222}Rn , ^{226}Ra and ^{238}U were 145.95 nSv/y and $132.44 \mu\text{Bq/kg}$ respectively. The results have shown all these parameters are within the permissible limits, and It turns out that the radioactivity almost has been present in all samples with a level recommended by UNSCEAR and ICRP. Therefore, the intake of the studied medical drugs does not lead to substantial changes in the internal effective dose.

Key words : ^{222}Rn , ^{226}Ra , ^{238}U , medical drugs, medical plants.

Introduction

Radium, radon and uranium are grouped together because they are radionuclides, unstable elements that emit ionizing radiation (Casarett and Doull, 2001). Ionizing radiation can cause toxicity when the particles pass into or through the body at high speed. If a collision occurs with the molecules of living cells, they may be damaged. These particular radionuclides emit radioactivity primarily in the form of alpha particles. Alpha radiation cannot pass through the dead outer layers of the skin (ATSDR, 1990). Therefore, these substances are a health risk only if taken into the body by ingestion or inhalation. Uranium, radium and radon occur naturally in the environment, uranium and radium as solids in rock while radon exists as a gas (ATSDR, 1990). The radioactive half-life is the time it takes for a substance to lose one-half of its radioactivity. While the half-life of radon is only about four days, the half-lives of the most common isotopes for radium and uranium, radium-226 and uranium-238, are approximately 1,600 years and 4.5 billion years, respectively (ATSDR, 1999). Generally, radionuclides are the source of the three types of the radiation are alpha

particles, beta particles and gamma rays (Lilley, 2001). The primary sources of elements from the environment to plants are: air, water and the soil (Chen *et al.*, 2005). The radionuclides present in the environment are transferred to plants by two ways : first indirect method uptake from soil through roots. When food crops are grown in the contaminated soil, the activity is shifted from the soil to the roots and then in shoots. At the end, activity is transferred to the human diet (Robinson, 1987). These radio nuclides can get transferred into plants along with the nutrients during mineral uptake and accumulate in various parts and even reach edible portions (Chen *et al.*, 2005). Second, it is a direct method absorption through aerial parts of the plants. Presence of radioactivity in plant organs has been reviewed by various workers (Robinson, 1987). Many medicinal plants are directly and indirectly used in the synthesis of medicines (Brown, 1992; Gurib-Fakim *et al.*, 2010). Amount of radionuclides accumulated in the medicines can be obtained by measuring their concentrations in them. The polluted-medicinal plants by radiation contribute to the increase of the internal effective dose (Vandenhove *et al.*, 2014). The elevated levels of radionuclides increase

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risks of developing lung cancer by emitting alpha particles. The elevated intake of radon, radium, and uranium in plants may result in hazardous effects in the human body (Bhatti and Malik, 1994). Therefore, the concentration measurement of these radionuclides in pharmaceuticals derived from medicinal plants is very important in order to evaluate the radiation dose and to prevent the exposure of consumers to radiation. The aim of this work was to estimate the concentrations of radon, radium and uranium in 22 samples of medical drugs (capsule) derived from medicinal plants, and collected from local Pharmacies in Iraq using LR-115 type II.

Materials and Methods

Sample collection and preparation

22 sample medical drugs (capsule) were collected from the local Pharmacies in Najaf, Iraq for the period from 1/10/2017 to 1/11/2017 as shown in Table 1.

Table 1 : Samples of medical drugs (capsule) samples under study.

No.	Sample name	Sample code
1	Vitalactib	H1
2	Swiss microlactin	H2
3	Uriclar	H3
4	Dizyme blend	H4
5	Legalon forte	H5
6	Actiloba plus	H6
7	Bran	H7
8	Acaiberry	H8
9	Speman	H9
10	Prostamed	H10
11	Ab slim extra	H11
12	Fat burner	H12
13	Digestives	H13
14	Ginseng	H14
15	Pilex	H15
16	Linaza	H16
17	Castor oil	H17
18	GR six	H18
19	Rowa chol	H19
20	Garlic	H20
21	Silymarin 140	H21
22	Ria Alwa	H22

After collecting all samples, the solid samples were dried in an oven at a temperature of 100°C for 5 hours. After that were crushed electronically, using electric mill. For homogeneity, the samples were sieved (0.8mm-pore-size sieve). The weights of the investigated samples were between 5 g and 18 g. Samples were placed at the bottom

of a sealed cylindrical plastic tube made of polyethylene, and were stored for about 1 month before counting to reach secular equilibrium between the isotopes of natural decay series.

Alpha particles measurement

ALR-115 Type-II with a thickness of (12µm) detector used the samples under study for measuring alpha particles levels. The detector was placed at the closed top end of a plastic cup diameter (3.5cm), length (7cm) and volume (130ml). Each piece was cut in area (1×1cm²) and put in the upper part of containers. A piece of the cut LR-115 type-II detector was placed at the bottom of each cylinder cover, with samples at the bottom of cylinder and then sealed for exposure of 62 day. The LR-115 type II detector has the capability to detect alpha particles emitted from radon and its progeny. Alpha particles make tracks when colliding with the detector. The number of tracks appropriate to the mean radon concentration. The exposed detectors were collected and etched chemically using 2.5 N NaOH at 60°C for 1.5 hour. After that, they were rinsed in distilled water. The track density on LR-115 type II (Tr/cm²) was counted using an optical microscope of magnification 10 × 40 (Hady *et al.*, 2016). The background correction was evaluated by subtracting background from registered alpha track density.

²²²Rn, ²²⁶Ra and ²³⁸U concentrations measurement

The concentration of radon (²²²Rn) in the airspace of the tube (C.) was calculated from the formula (Mayya *et al.*, 1998):

$$C.B_q = \rho \frac{E_m F K t}{\dots} \quad (E1F)$$

Where, ρ is the track density on the exposed detector (Tr/cm²), t is the exposure time of the sample (62 d) and K is the diffusion constant (calibration factor or sensitivity factor). K was determined from the following relationship using the values of the tube (Barillon *et al.*, 1993):

$$K = 0.24 r_E 2 \cos \theta - r' F \quad (E2F)$$

Where, r is radius of the tube (r = 1.75cm), θc is the critical angle of the LR-115 type II detector (θc = 40°) (Barillon *et al.*, 1993) and r_a is the range of alpha particle in air (r_a = 4.15 cm) (Fleischer and Mogro-Champero, 1978). Consequently, the diffusion constant is 0.04 (Tr./cm²d) per (Bq/m³).

Can be calculated radon concentration within the sample (C.), the following relation was used (Elzain,

2014):

$$C \cdot \frac{Bq}{E_{nf} F} = C \lambda_{Rn} h t \quad (E3F)$$

Where, λ_{Rn} is the decay constant of ^{222}Rn (0.1814 d^{-1}), h is the distance from the surface of the sample to the detector, t is the exposure time (62 d) and l is the thickness of the sample in the tube.

The activity concentration of radon inside sample (C) was determined using the relation :

$$C \cdot \frac{Bq}{E_{kg} F} = \frac{C I A}{M} \quad (E4F)$$

Where, A^s is the surface area of the sample and M^s is the mass of the investigated sample.

The activity concentration of radium (^{226}Ra) within the sample (C) was determined using the relationship (Azam *et al.*, 1995):

$$C \cdot \frac{Bq}{E_{kg} F} = \frac{C h A}{M} \quad (E5F)$$

The radon activity inside the sample (A) was obtained using the following formulas :

$$A \cdot \frac{Bq}{E_{qF}} = C \cdot V \quad (E6F)$$

$$V = \pi l r \quad (E7F)$$

Where, V^s is the sample volume in m^3 .

The number of uranium (^{238}U) atoms in the sample (N) at the secular equilibrium can be obtained by Podgorsak (2005):

$$N = \frac{A}{\lambda} \quad (E8F)$$

Where, λ_U is the decay constant of uranium ($4.9 \times 10^{18}/\text{s}$). Therefore, the weight of uranium in the sample (M) in gram can be determined as following (Wong, 2008):

$$M = \frac{N A}{N_A} \quad (E9F)$$

Where, A_U is the mass number of ^{238}U and N_A is Avogadro's number. Thus, the concentration of uranium (C) in ppm is given by :

$$C \cdot \frac{E_{ppm} F}{M} = \frac{M}{M} \quad (E10F)$$

Annual average internal dose AAIED and risk of an excess cancer fatality per million person

(RECFMP)

The annual average internal dose by an ingestion of radionuclides has been calculated according to the equation (Al-Omari, 2015) :

$$AAIED \frac{E^n \text{Sv}_F}{y} = C \times I \times C \quad (E11F)$$

Where, I^m is the consumption rate from the intake of the medicine for person in one year (kg/y) and C is the effective dose conversion factor of the radioactive element (nSv/Bq). In evaluating the AAIED due to radionuclides ingestion from the intake of medicines, the calculations were based on assuming a twice-daily consuming of 500 mg for each medicine (Al-Omari, 2015). While, the effective dose conversion factor for radon, radium and uranium ingestion by people as 3.5 nSv/Bq (NRC, 1999), 960 nSv/Bq (Ahmed, 2004) and 45 nSv/Bq (ICRP, 1995). The risk of an excess cancer fatality per million person due to ingestion of radon, radium, and uranium from pharmaceuticals has been calculated according to the equation (Ali Abid *et al.*, 2017):

$$RECFMP = AAIED \times DL \times RF \quad (E12F)$$

Where, DL is the duration of life (70 yr) and RF is the risk factor (0.055 Sv⁻¹) recommended by the ICRP (Clarke and Bines, 2011).

Results and Discussion

The mean results of arithmetic for ^{222}Rn , ^{226}Ra and ^{238}U concentrations in some samples of medical drugs (capsule) in the present study have been given in table 2. From table 2 and fig. 1, the minimum value of (C) was for sample H15 ($0.51 \text{ Bq}/\text{m}^3$), while the maximum value was found in pharmaceutical H18 ($60.16 \text{ Bq}/\text{m}^3$) with an average value of $16.78 \text{ Bq}/\text{m}^3$. The average value of (C) and (C) was $1038.01 \text{ Bq}/\text{m}^3$ and $1.64 \text{ mBq}/\text{kg}$, respectively (table 2). The variation in the radon concentrations may be due to the various ingredients of these pharmaceuticals because they were of plant origin. The pollution by radiation may be also directly caused by the absorption of radionuclides from the atmosphere. The plant uptake of radionuclides is different depending on the crust of the soil, the plant itself and the fertilizer. Consequently, the radiation pollution of the plant is expected. However, the radon concentrations for all of the investigated pharmaceuticals were much less than acceptable lower limit of the action level, which is 200–600 Bq/m^3 (Al-Omari, 2015). Upon comparing the values of (C) and (C) for all of the investigated formulations (table 1), it is obvious that the radon concentration inside the sample is much greater than that in the airspace of

Table 2 : Results of Radon, Radium and Uranium in Drugs (capsule) samples under study.

No.	Sample code	²²² Rn			²²⁶ Ra	²³⁸ U
		C.(Bq/m ³)	C.(Bq/m ³)	C.(Bq/kg)	C.(mBq/kg)	C.(μBq/kg)
1	H1	0.52	31.67	0.06	5.42	0.06
2	H2	10.24	633.46	1.22	108.32	1.21
3	H3	42.24	2613.02	3.59	319.17	3.58
4	H4	4.61	285.06	0.34	30.47	0.34
5	H5	32.00	1979.56	4.76	423.14	4.74
6	H6	2.30	142.53	0.27	24.37	0.27
7	H7	17.92	1108.56	2.13	189.57	2.12
8	H8	7.68	475.10	0.33	29.02	0.33
9	H9	15.36	950.19	0.54	47.79	0.54
10	H10	0.78	47.51	0.03	2.54	0.03
11	H11	8.70	538.44	0.65	57.55	0.64
12	H12	11.01	680.97	0.41	36.39	0.41
13	H13	55.04	3404.85	2.34	207.94	2.33
14	H14	2.56	158.37	0.12	10.42	0.12
15	H15	0.51	31.67	0.02	1.69	0.02
16	H16	4.35	269.22	0.18	16.44	0.18
17	H17	14.59	902.68	0.96	85.76	0.96
18	H18	60.16	3721.58	8.95	795.51	8.91
19	H19	0.77	47.51	0.23	20.31	0.23
20	H20	17.15	1061.05	2.04	181.44	2.03
21	H21	57.35	3547.38	6.82	606.62	6.80
22	H22	3.33	205.87	0.12	11.00	0.12
Average		16.78	1038.01	1.64	145.95	1.64

the tube. The large difference between the two values may be attributed to the fact that most of the generated radon atoms inside sample decay before entering the space of the tube since they have low decay time of 3.82 d (Al-Omari, 2014, 2015) as well as due to the existence of the radon parent (radium) inside the sample, but not in the airspace of the tube.

The value of activity concentration of radium (*C.*) were between 1.69 mBq/kg and 795.51 mBq/kg with an average of 145.95 mBq/kg (table 2). The variation of the activity concentration of radium in the formulations may be attributed to the fact that the activity concentrations of radium and other radionuclides vary from one soil of cultivation to another, as well as plants differ in their uptake of radionuclides. Plants may be polluted through root uptake, direct deposition, fertilizer, and irrigation with contaminated water. As from table 2, alpha activities due to radium in the medicine samples are lower than those due to radon. This is because radon has less half- life (3.82 d) than radium (1600 y) (Elzain, 2014; Ali Abid *et al.*, 2017).

C. varied from 0.02 μBq/kg in sample H15 to 8.91 μBq/kg in sample H18 with an average value of 1.64

μBq/kg. Therefore, the activity concentration of radon (table 1) is higher than that of uranium and hence uranium contribution to the alpha particles emission is negligible. However, the concentrations of radionuclides in the current study are much less than those of medicinal plants (Desideri *et al.*, 2010). This is because, during the preparation processes, the activity concentrations of radionuclides are significantly reduced in medicinal plant formulations compared to those in the raw plants. The variation in concentrations is probably due to the different natural existence of uranium in most plants.

There is a positive correlation of 0.894 between radon concentration in the airspace of the tube and radium activity concentration (fig. 2). This indicates that the origin of the radon is due to the radium in the sample since radon concentration depends on radium content (Al-Omari, 2015; Yalim *et al.*, 2007) and this relation can be used as a ruler to find the concentration of radon or radium for the current samples.

Table 3 shows the annual average internal effective dose (AAIED) and risk of an excess cancer fatality per million person (RECFPMP) due to ingestion of radon, radium, and uranium from samples of medical Drugs

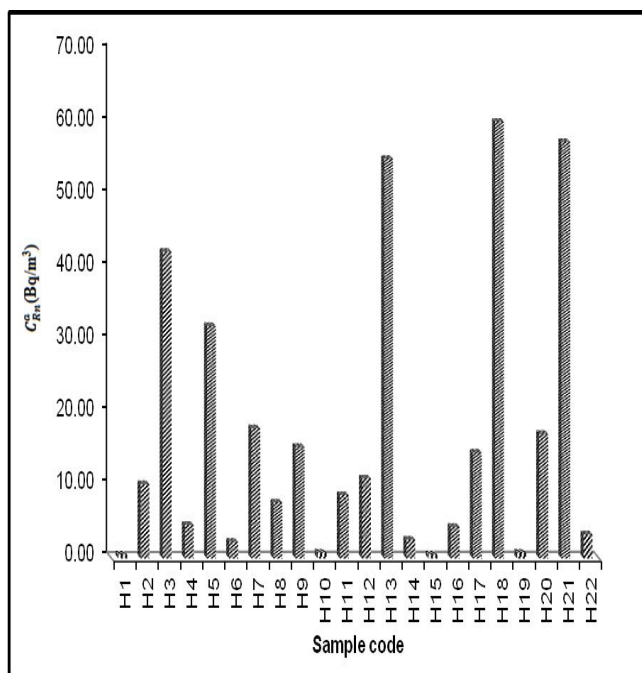


Fig. 1 : Radon concentration the airspace above the sample.

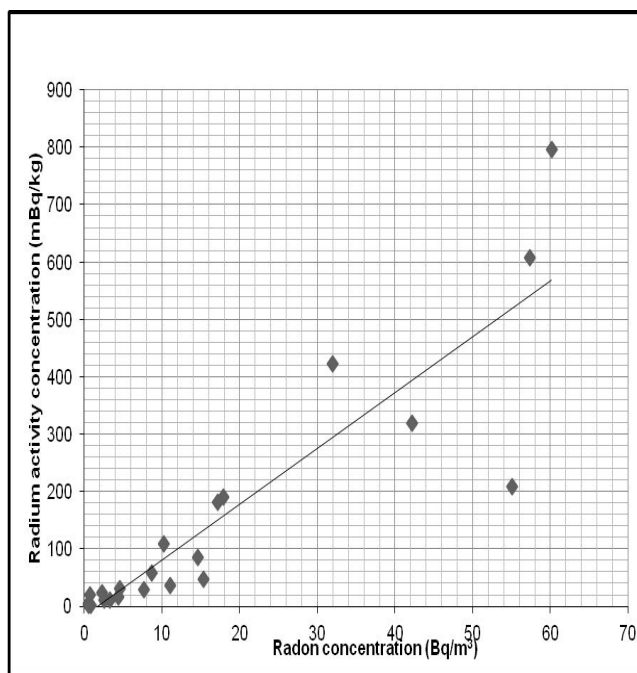


Fig. 2 : Correlation between (C.) and (C.)

Table 3 : Results of AAIED and RECFPMP in Drugs (capsule) samples under study.

No.	Sample code	AAIED (nSv/y)			Total AAIED (nSv/y)	RECFPMP
		²²² Rn	²²⁶ Ra	²³⁸ U		
1	H1	0.08	1.90	0.000001	1.98	0.008
2	H2	1.56	37.96	0.000020	39.51	0.152
3	H3	4.59	111.84	0.000059	116.42	0.448
4	H4	0.43	10.68	0.000006	11.11	0.043
5	H5	6.08	148.27	0.000078	154.35	0.594
6	H6	0.34	8.54	0.000004	8.88	0.034
7	H7	2.72	66.43	0.000035	69.15	0.266
8	H8	0.42	10.17	0.000005	10.59	0.041
9	H9	0.69	16.75	0.000009	17.44	0.067
10	H10	0.04	0.89	0.000000	0.93	0.004
11	H11	0.83	20.17	0.000011	21.00	0.081
12	H12	0.52	12.75	0.000007	13.27	0.051
13	H13	2.99	72.86	0.000038	75.85	0.292
14	H14	0.15	3.65	0.000002	3.80	0.015
15	H15	0.03	0.59	0.000000	0.62	0.002
16	H16	0.23	5.76	0.000003	5.99	0.023
17	H17	1.23	30.05	0.000016	31.28	0.120
18	H18	11.43	278.75	0.000146	290.18	1.117
19	H19	0.29	7.12	0.000004	7.41	0.029
20	H20	2.61	63.58	0.000033	66.18	0.255
21	H21	8.71	212.56	0.000112	221.27	0.852
22	H22	0.15	3.85	0.000002	4.01	0.015
Average		2.10	51.14	0.000027	53.24	0.20

(capsule) derived from medical plants. From table 3, it is found that the average value of AAIED by ingestion of radon, radium, and uranium in samples under study are 2.10 nSv/y, 51.14 nSv/y and 0.000027 nSv/y, respectively. Also, from table 3, the total AAIED was between 0.62 and 290.18 nSv/y with an average value of 53.24 nSv/y. The value of RECFPMP ranged between 0.002 and 1.117 with an average value of 0.20. Therefore, the maximum AAIED of H18 is much smaller than the UNSCEAR recommended safety limit for public, which is 0.29 mSv/y for the ingestion exposure caused by natural sources (UNSCEAR, 2000). Further, the AAIED value of radon in sample H18 is also very low compared to the action level of 3–10 mSv/y recommended by International Commission on Radiological Protection (Field, 1999). Hence, the intake of the current medicines is safe from the viewpoint of radiation pollution.

Conclusion

^{222}Rn , ^{226}Ra and ^{238}U concentrations in some samples of medical drugs (capsule) derived from medical plants in Iraq have analyzed successfully. Average concentrations of radon, radium, and uranium in 22 samples were much smaller than the average world level. The average of the total annual internal effective doses from radon, radium, and uranium due to ingestion of all samples in the present study was lower than the action levels recommended by UNSCEAR and ICRP. As a result, the intake of the current pharmaceuticals does not result insignificant changes in the internal radiation dose.

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