Radioactivity measurement of ^{222}Rn, ^{226}Ra and ^{238}U in pharmaceuticals and evaluation of cancer risk

Saleh Al-Omari

Department of Physics,
Faculty of Science,
The Hashemite University,
Zarqa 13115, Jordan
Email: salomar@hu.edu.jo

Abstract: The concentrations of ^{222}Rn, ^{226}Ra, and ^{238}U were measured in 16 pharmaceuticals derived from medicinal plants using a solid-state nuclear track detector (CR-39). The radon levels in the airspace above the samples ranged between 0.04 and 20 Bq/m^3 with an average value of 5.82 Bq/m^3, while the uranium concentration ranged from 0.04 to 25.62 mppm with an average value of 7.34 mppm. The activity concentrations for radon, radium, and uranium were 0.53–344.4 mBq/kg, 0.05–30.63 mBq/kg, and 0.53–344.4 μBq/kg, respectively. The maximum annual average internal effective dose from radon, radium, and uranium due to a twice-daily ingestion of 500-mg tablet for one of the investigated medicines was 11.18 nSv/y. This value is much smaller than the UNSCEAR recommended safety limit for public, which is 290,000 nSv/y for the ingestion exposure caused by natural sources. Therefore, the intake of the studied pharmaceuticals does not lead to substantial changes in the internal effective dose.

Keywords: radon; radium; uranium; solid-state nuclear track detector; CR-39; pharmaceutical; internal effective dose.

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Biographical notes: Saleh Al-Omari is an Associate Professor of biophysics at the Physics Department of the Hashemite University, Jordan. He obtained his PhD in biophysics from Berlin University, Germany. He is also interested in radiological researches including WDXRF, EDXRF, and radon measurements. He has taught several undergraduate and graduate courses including radiation physics and supervised many graduate students in the field of XRF. He has published many scientific papers in international journals.

1 Introduction

Uranium (^{238}U) is one of the natural constituents of the crust of Earth and its presence in soil is naturally causing radiation pollution (Newsome et al., 2014). The different industrial sources and agricultural activities are also responsible for the soil pollution by

uranium (Luo and Gu, 2011). The distribution of uranium is generally affected by different parameters such as climate, mineralogy, type of main source, relative mobility, and sort of soil (Williams et al., 2013; Schery et al., 1989).

Inert radon gas (^{222}Rn) is generated from the radioactive decay of radium (^{226}Ra) in the uranium transformation decay in the crust of the Earth (UNSCEAR, 2000). Radium will then decay into radon gas that has a half-life time of 3.8 d (Al-Omari, 2014). Radon and its airborne decay products are considered the most risky radioactive elements that are responsible for the internal health danger.

Uranium, which is the main source of radon, may enter into the plants, foods, and drinking water through natural processes and/or because of human activities (Shiraishi et al., 2000; UNSCEAR, 2000; INFOSAN, 2011). Uranium can cause damage to the waste filtering and disposal system of the body. The radionuclides uptake in the plant depends on type of plant (Kozhakhanov et al., 2014). Many medicinal plants are directly and indirectly used in the synthesis of medicines (Brown, 1992; Gurib-Fakim and Kasilo, 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 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 the present study was to determine concentrations of radon, radium, and uranium in 16 formulations derived from medicinal plants, and collected from local stores in Jordan, were carried out using Solid-State Nuclear Track Detector (SSNTD) technique. This method is widely used as a detection tool to obtain the tracks of alpha particles and subsequently the radon concentration (Al-Omari, 2014). The activity concentration, the Annual Average Internal Effective Dose (AAIED), and radiological risk from ingestion of these medicines due to radon, radium, and uranium were also evaluated. The results have been compared with the limits of international recommended values of safety standards.

2 Experimental methods

2.1 Sampling

A total of 16 pharmaceuticals of different medicinal plants were purchased from local stores in Jordan. The samples were dried in an oven at a temperature of 95°C for 20 hours. After that, they were crushed, homogenised, and sieved to about 60 micron-mesh size to obtain fine powder of similar particle sizes. The weights of the investigated samples were between 9.25 and 27.37 g (Table 1). Samples were placed at the bottom of a sealed cylindrical plastic tube made of polyethylene.

Table 1 Sample code, sample mass (M^s), sample thickness (l), concentration of radon in the airspace of the tube (C^a_{Rn}), radon concentration inside sample (C^s_{Rn}), activity concentration of radon inside sample ($C^{s,ac}_{Rn}$), activity concentration of radium inside sample ($C^{s,ac}_{Ra}$), activity of radon inside sample (A^s_{Rn}), number of uranium atoms in sample (N^s_U), weight of uranium in sample (M^s_U), concentration of uranium (C^s_U) in sample, and activity concentration of uranium inside sample ($C^{s,ac}_U$)

Sample code	M^s *(g)*	l *(cm)*	C^a_{Rn} *(Bq/m³)*	C^s_{Rn} *(Bq/m³)*	$\frac{C^s_{Rn}}{C^a_{Rn}}$	$C^{s,ac}_{Ra}$ *(mBq/kg)*	$C^{s,ac}_{Rn}$ *(mBq/kg)*	A^s_{Rn} *(mBq)*	$N^s_U \times 10^{14}$	M^s_U *(μg)*	C^s_U *(mppm)*	$C^{s,ac}_U$ *(μBq/kg)*
S_1	27.37	4	8	134.96	16.9	81.91	7.28	2.242	4.58	0.18	6.58	81.91
S_2	17.41	4.2	6	93.19	15.5	93.36	7.44	1.625	3.32	0.13	7.47	93.36
S_3	22.15	3.9	2	35.18	17.6	25.72	2.29	0.570	1.16	0.046	2.08	25.72
S_4	20.11	4.3	1	14.91	14.9	13.24	1.18	0.266	0.54	0.021	1.04	13.24
S_5	21.03	4.1	0.04	0.65	16.3	0.53	0.05	0.011	0.02	0.001	0.04	0.53
S_6	20.50	3.7	5	95.75	19.2	71.76	6.38	1.471	3.00	0.12	5.85	71.76
S_7	17.17	3.8	5	91.75	18.4	84.32	7.5	1.449	3.06	0.12	7.09	84.32
S_8	19.00	4	15	253.05	16.9	221.23	19.67	4.203	8.58	0.34	17.9	221.23
S_9	18.41	3.6	2	39.99	20	32.47	2.89	0.598	1.22	0.048	2.61	32.47
S_{10}	10.52	4.4	4	57.26	14.3	99.45	8.84	1.046	2.13	0.084	7.98	99.45
S_{11}	16.00	4.1	20	323.69	16.2	344.40	30.63	5.111	10.43	0.41	25.62	344.40
S_{12}	21.00	3.8	3	55.05	18.35	41.37	3.68	0.869	1.77	0.070	3.33	41.37
S_{13}	18.76	4.3	7	104.36	14.9	99.33	8.83	1.863	3.80	0.15	7.80	99.33
S_{14}	21.38	3.5	3	62.66	20.9	42.60	3.79	0.911	1.86	0.074	3.46	42.60
S_{15}	19.58	4.2	8	124.25	15.5	110.68	9.84	2.167	4.42	0.17	8.68	110.68
S_{16}	9.25	3.9	4	70.36	17.6	123.19	10.95	1.140	2.33	0.092	9.95	123.19
Min–max	9.25–27.37	3.5–4.4	0.04–20	0.65–323.69	14.3-20.9	0.53–344.4	0.05–30.63	0.011–5.111	0.02–10.43	0.0008–0.41	0.04–25.62	0.53–344.4
Average	18.7	4	5.8	97.3	17.1	93	8.20	1.6	3.1	0.128	7.34	93

2.2 Alpha particles measurement

SSNTD of CR-39 was cut into pieces of area 1 × 1 cm^2 and fixed with double-sided adhesive tape to the inner surface of the top cover of the sealed cylindrical plastic. The sensitive side of CR-39 was directed downward and above the sample. The radius of the cylindrical tube and its height were 1.15 and 10 cm, respectively. This arrangement agrees with the recommended dimensions of the tube that give a considerable confidence in radon measurements (Nikezic et al., 1996). Background on the films was estimated by placing these detectors for 62 d in empty well-closed plastic tubes as the same of those used for analysing the samples and counting the produced track densities. This process was repeated four times. The sealed tubes including samples were then stored for 62 d. Because the exposure time was 62 d and the system is well sealed, there is no escape of radon. Therefore, it was assumed secular radioactive equilibrium between ^{226}Ra and each of its decay products in the uranium series.

The CR-39 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 appropriates to the mean radon concentration. The exposed detectors were collected and etched chemically using 6.25 M NaOH at 75°C for 7 hours. After that, they were rinsed in distilled water. The track density on CR-39 (Tr/cm^2) was counted using an optical microscope of magnification 10 × 40 (Al-Omari, 2014). The background correction was evaluated by subtracting background from registered alpha track density.

2.3 Radon concentration measurement

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

$$C^a_{Rn} = \rho t^{-1} K^{-1} \tag{1}$$

where ρ is the track density on the exposed detector (Tr/cm^2), t is the exposure time of the sample (62 d), and K is the diffusion constant (calibration factor or sensitivity factor). The last parameter was determined from the following relationship using the values of the tube (Barillon et al., 1993):

$$K = 0.25r\left(2\cos\theta_c - r r_\alpha^{-1}\right) \tag{2}$$

where r is radius of the tube (r = 1.15 cm), θ_c is the critical angle of the CR-39 detector (θ_c = 35°) (Barillon et al., 1993), and r_α is the range of alpha particle in air (r_α = 4.15 cm) (Fleischer et al., 1978). Consequently, the diffusion constant is 0.03387 Tr/cm^2/d/Bq m^3.

In order to obtain radon concentration within the sample, the following relation was used (Elzain, 2014):

$$C^s_{Rn} = \lambda_{Rn} C^a_{Rn} h t l^{-1} \tag{3}$$

where C^s_{Rn} is the radon concentration inside the sample (Bq/m^3), λ_{Rn} is the decay constant of ^{222}Rn (0.1814/d), 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 ($C_{\mathrm{Rn}}^{s,ac}$) in Bq/kg was determined using the relation:

$$C_{\mathrm{Rn}}^{s,ac} = C_{\mathrm{Rn}}^{s} l A^{s} M^{s-1} \tag{4}$$

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 ($C_{\mathrm{Rn}}^{s,ac}$) in Bq/kg was determined using the relationship (Azam et al., 1995):

$$C_{\mathrm{Ra}}^{s,ac} = C_{\mathrm{Rn}}^{a} h A^{s} M^{s-1} \tag{5}$$

The radon activity inside the sample (A_{Rn}^{s}) was obtained using the following formulas:

$$A_{\mathrm{Rn}}^{s} = C_{\mathrm{Rn}}^{s} V^{s} \tag{6}$$

$$V^{s} = \pi l r^{2} \tag{7}$$

where V^s is the sample volume in m^3.

2.4 Uranium concentration measurement

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

$$N_U^s = \lambda_U^{-1} A_{\mathrm{Rn}}^{s} \tag{8}$$

where λ_U is the decay constant of uranium (4.9×10^{-18}/s). Therefore, the weight of uranium in the sample (M_U^s) in gram can be determined as following (Samuel, 2004):

$$M_U^s = \frac{N_U^s A_U}{N_A} \tag{9}$$

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

$$C_U^s\,(\mathrm{ppm}) = \frac{M_U^s}{M^s}. \tag{10}$$

3 Results

In order to determine the level of radiation pollution of the 16 medicinal plant pharmaceuticals, it was obtained ^{222}Rn concentration in Bq/m^3 in the airspace of the test tube as well as ^{238}U concentration in the sample in ppm as an assessment for this possible pollution. The detailed results for the determination of the radiation pollution using solid-state nuclear track detection method have been listed in Table 1. Activity concentration inside sample for radon ($C_{\mathrm{Rn}}^{s,ac}$), radium ($C_{\mathrm{Ra}}^{s,ac}$), and uranium ($C_U^{s,ac}$) was 0.53–344.4 mBq/kg, 0.05–30.63 mBq/kg, and 0.53–344.4 μBq/kg, respectively.

Table 2 shows the AAIED in nSv/y and risk of an excess cancer fatality per million person (RECFPMP) due to ingestion of radon, radium, and uranium from pharmaceuticals. The total AAIED was between 0.018 and 11.178 nSv/y with an average value of 2.994 nSv/y. The value of RECFPMP ranged between 0.00018 and 0.11178 with an average value of 0.02994.

Table 2 Estimate of annual average internal effective dose (AAIED) in nSv/y and risk of an excess cancer fatality per million person (RECFPMP) due to ingestion of ^{222}Rn, ^{226}Ra, and ^{238}U from formulations derived from medicinal plants

Sample code	*AAIED (nSv/y)*			*Total AAIED (nSv/y)*	*RECFPMP*
	^{222}Rn	^{226}Ra	^{238}U		
S_1	0.104	2.5509	0.0013454	2.656	0.02656
S_2	0.119	2.6069	0.001533	2.727	0.02727
S_3	0.03289	0.802	0.0004225	0.835	0.00835
S_4	0.0169	0.41347	0.0002175	0.431	0.00431
S_5	0.000677	0.01752	0.0000087	0.018	0.00018
S_6	0.0916	2.23555	0.0011786	2.328	0.02328
S_7	0.1077	2.628	0.0013849	2.737	0.02737
S_8	0.2826	6.8923	0.0036337	7.179	0.07179
S_9	0.0414	1.0126	0.0005333	1.055	0.01055
S_{10}	0.127	3.0975	0.001633	3.226	0.03226
S_{11}	0.4399	10.732	0.0056567	11.178	0.11178
S_{12}	0.05285	1.289	0.0006795	1.343	0.01343
S_{13}	0.12689	3.0940	0.0016314	3.223	0.03223
S_{14}	0.054	1.3280	0.0006997	1.383	0.01383
S_{15}	0.14139	3.4479	0.0018179	3.591	0.03591
S_{16}	0.15737	3.83688	0.0020233	3.996	0.03996
Min–max	0.000677–0.4399	0.01752–10.732	0.0000087–0.00566	0.018–11.178	0.00018–0.11178
Average	0.11851	2.87403	0.00152	2.994	0.02994

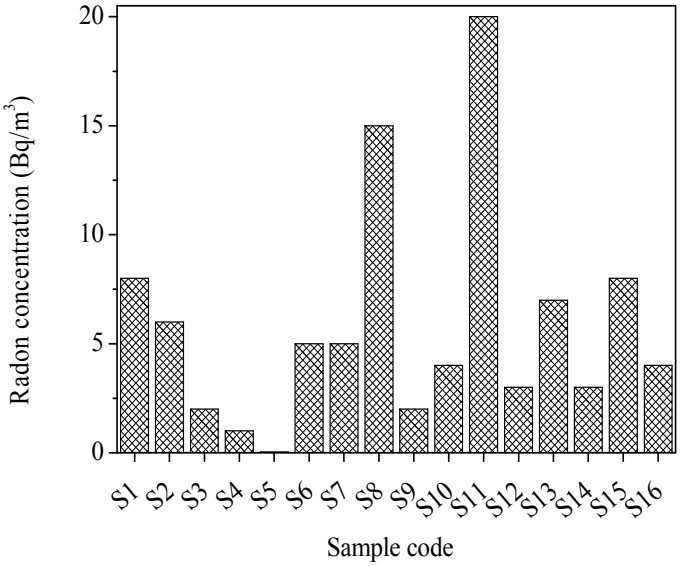
4 Discussion

4.1 Activity concentration

As presented in Table 1 and Figure 1, the least radon concentration in the airspace of the tube (C^a_{Rn}) was for sample S_5 (0.04 Bq/m^3), while the maximum value was found in pharmaceutical S_{11} (20 Bq/m^3) with an average value of 5.815 Bq/m^3. Furthermore, sample S_8 has relatively high radon concentration of 15 Bq/m^3. Rest of the pharmaceuticals have comparable radon concentrations. 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 fertiliser.

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, 2014).

Figure 1 Radon concentration (Bq/m^3) in the airspace above the sample



The radon concentration inside the sample (C_{Rn}^{s}) is between 0.65 and 323.69 Bq/m^3 with an average value of 97.3 Bq/m^3 (Table 1). Upon comparing the values of C_{Rn}^{s} and C_{Rn}^{a} 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 the tube by a factor ranged between 14 and 21 with an average value of about 17. 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), as well as due to the existence of the radon parent (radium) inside the sample, but not in the airspace of the tube. Consequently, $C_{\mathrm{Rn}}^{s} \gg C_{\mathrm{Rn}}^{a}$.

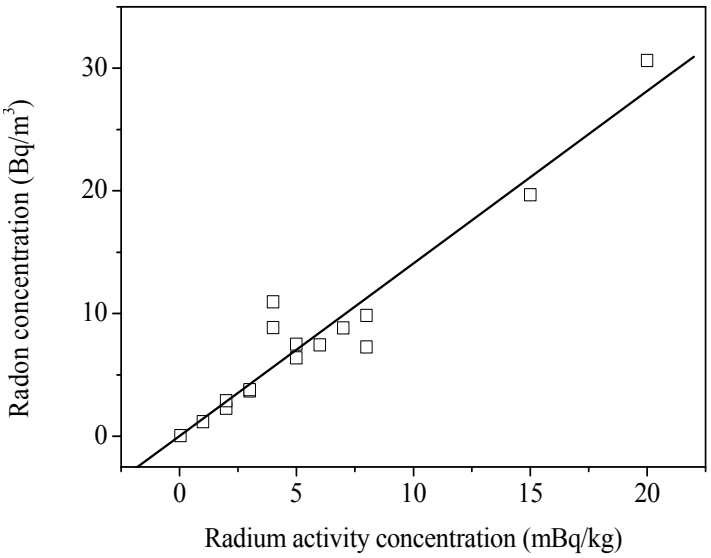
To obtain number of radon alpha particles emitted per second inside the medicine, the value of $C_{\mathrm{Rn}}^{s,ac}$ has been used. It ranged between 0.53 mBq/kg for sample S_5 and 344.4 mBq/kg for sample S_{11} with an average value of 93 mBq/kg. This means that for the pharmaceutical S_{11} of 500-mg tablet, which has the maximum activity concentration, its activity is 0.172 mBq. In other words, one alpha particle originated from radon is emitted every 96.8 min. Assuming two days are needed for the excretion of this medicine from the body, the deposited alpha particles before the excretion are about 30. On the other hand, the minimum value of $C_{\mathrm{Rn}}^{s,ac}$ was found in sample S_5 ($C_{\mathrm{Rn}}^{s,ac}$ = 0.53 mBq/kg). Therefore, 500-mg tablet of this pharmaceutical gives an activity of 0.265 μBq. This means that one alpha particle is emitted every 43.7 d. Considering two days are needed for the excretion of the pharmaceutical S_5 from the body, the probability of deposited one alpha particle in the body before excretion is 4.6%. Therefore, from the viewpoint of radiation pollution, the medicine S_5 is the safest in the current study.

Values of $C_{\mathrm{Ra}}^{s,ac}$ were between 0.05 and 30.63 mBq/kg with an average of 8.203 mBq/kg. 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, fertiliser, and irrigation with contaminated water. As seen from Table 1, alpha activities due to radium in the medicine samples are lower than those due to radon. On average, activity concentration of radon was about 11 times higher than that of radium. This is because radon has less half life (3.82 d) than radium (1600 y).

There is a positive correlation of 0.9600 between radon concentration in the airspace of the tube and radium activity concentration (Figure 2). This indicates that the origin of the radon is due to the radium in the sample since radon concentration depends on radium content (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.

The determination of uranium amount in pharmaceuticals is important owing to its radiation exposure and chemical toxicity (Kurttio et al., 2002). The main health effect of uranium is attributed to the chemical kidney toxicity leading to kidney failure by preventing the normal disposal of wastes. About 67% of the uranium transported to the blood is filtered by the kidney and excreted in the urine during 24 hours. Roughly, 90 μg of uranium exists in the human body from usual intakes of air, food, and water. Upon occurrence of the damage of the human kidneys, quantity of uranium exceeds 90 μg resulting in carcinogenic effects (Somogyi, 1986).

Figure 2 Correlation of radon concentration (Bq/m^3) and radium activity concentration (mBq/kg)

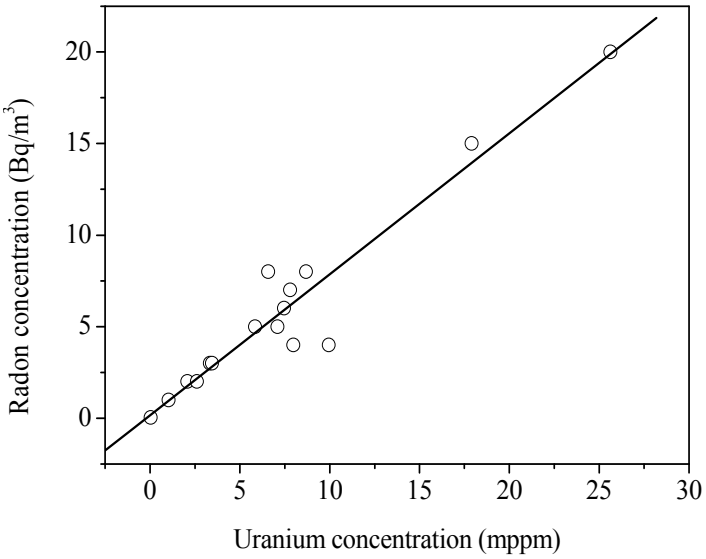


Uranium concentration varied from 0.04 mppm in sample S_5 to 25.62 mppm in sample S_{11} with an average value of 7.34 mppm. The variation in concentrations is probably due to the different natural existence of uranium in most plants. The daily ingestion of natural uranium through food and water is about 1–2 μg and 1.5 μg/l, respectively (Singh and

Sinha, 2012). Assuming water consumption of the human as 2 l/d, the intake of the human through the pathway of water drink is 2 × 1.5 μg. Therefore, the total daily ingestion of uranium through food and water is 4–5 μg. If the daily consumption of sample S_{11}, which has the maximal concentration, is two 500-mg tablets, then the maximal contribution of S_{11} to the daily uranium intake is 25.62 ng. This value is much less than the total daily ingestion of uranium, which is 4–5 μg and does not exceed the limiting value of 90 μg which results in cancer. Therefore, the intake of the current medicines is secure and does not elevate significantly the daily dosage of uranium.

Figure 3 shows a strong linear positive correlation factor of 0.9604 between radon concentration in the airspace of the tube and uranium concentration. This means that uranium and subsequently radium are main pathways of radon generation for these pharmaceuticals. Furthermore, this strong correlation allows to obtain radon concentration if the uranium concentration is known and vice versa.

Figure 3 Correlation of radon concentration (Bq/m^3) and uranium concentration (mppm)



Values of $C_U^{s,ac}$ ranged from 0.53 to 344.4 μBq/kg with an average value of 93 μBq/kg. Therefore, the activity concentration of radon (Table 1) is 1000 times 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.

4.2 Annual average internal dose by an ingestion of radionuclides

The AAIED in nSv/y has been calculated according to the equation:

$$\mathrm{AAIED}\,(\mathrm{nSv}/y) = C^{s,ac} \times I^m \times C^f \left(\mathrm{nSv}/\mathrm{Bq}\right) \tag{11}$$

where $C^{s,ac}$ is the activity concentration of the radioactive element, I^m is the consumption rate from the intake of the medicine for person in one year (kg/y), and C^f 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. Since medicine S_{11} has maximal activities (Table 1), it will show the limit of the maximum AAIED. Therefore, it was presented as an example to demonstrate the calculation. Of course, the rest of the investigated pharmaceuticals have lower values compared to S_{11} (Table 1) because they have smaller radionuclides levels (Table 1).

The received values of AAIED by ingestion of radon, radium, and uranium in medicines are summarised in Table 2. Regarding the contribution of radon to the maximum AAIED, the annual accumulation amount of pharmaceutical S_{11} due to the daily intake of double 500-mg tablets is 365 g/y. Therefore, taking the radon activity inside pharmaceutical S_{11} as $C_{\mathrm{Rn}}^{s,ac}$ = 344.40 mBq/kg (Table 1) results in an annual activity intake of 0.1257 Bq. Moreover, applying the ingestion dose coefficient of radon as 3.5 nSv/Bq (NRC, 1999), which is applied for the ingestion of water, the resulting AAIED would be only 0.4399 nSv/y (Table 2). The overall contribution of radon from all of the samples to the average AAIED was 0.11851 nSv/y (Table 2). Therefore, the maximum AAIED of S_{11} 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 S_{11} is also very low compared to the action level of 3–10 mSv/y recommended by International Commission on Radiological Protection (ICRP, 1993). Hence, the intake of the current medicines is safe from the viewpoint of radiation pollution.

AAIED value of radium was also estimated by considering the daily intake of two 500-mg tablets, which result in a yearly consumption rate of 365 g. Therefore, the maximum activity concentration of radium that belongs to pharmaceutical S_{11} (Table 1) results in a yearly intake of 11.18 mBq. Taking the dose conversion factor for radium ingestion by people as 960 nSv/Bq (Ahmed, 2004), the maximum value of AAIED applying equation (11) is 10.7 nSv/y. The average contribution of radium from all of the medicines to the AAIED was 2.87 nSv/y (Table 2). These limits are much smaller than the recommended standards as mentioned above; hence, the medicines intake does not lead importantly to the increase in the internal effective dose. As obtained (Table 2), the overall average internal effective dose of radium for the current medicines is about 24 times higher than that of radon.

Assuming a twice-daily consumption of the medicine 500 mg, the accumulation amount of each pharmaceutical is 365 g/y. Therefore, the maximal contribution of uranium to the yearly internal activity due to ingestion of medicine S_{11} is 0.1257 Bq. Using the conversion factor for ingestion dose of uranium as 45 nSv/Bq (ICRP, 1995) and applying equation (11), the resulting internal effective dose is only 5.6 nSv/y. Again, this upper limit is much below the standards of UNSCEAR and ICRP. The average contribution of medicines to the AAIED of uranium is 0.0015 nSv/y (Table 2) which is about 1/78 of radon inside the samples. In addition, this value is smaller than the average world values for ingestion (between 0.2 and 0.8 mSv) (UNSCEAR, 2000).

Overall, the three radioisotopes of radon, radium, and uranium induced total maximum AAIED in sample S_{11} as 11.18 nSv (Table 2). This upper dose was below permissible dose limit of the ICRP (1 mSv/y) from all pathways (ICRP, 2005). Since

there is a risk per 0.1 Sv of the effective dose due to radionuclide exposures (NAS, 1990), the net upper dose of medicine S_{11} corresponds to risk of an excess cancer fatality per million persons of 0.11 in a million (Table 2), which is much below the US Environmental Protection Agency's guideline of 10 – 4 in a million (USEPA, 2006). Furthermore, the AAIED of other medicines in the current study due to ingestion of natural radionuclides is also far below the average radiation dose of 0.3 mSv/y (UNSCEAR, 2000). The total annual average global exposure to natural radiation sources is 2.4 mSv (Choi et al., 2008); therefore, the total maximum value of AAIED for pharmaceutical S_{11} due to ingestion of radon, radium, and uranium (11.18 nSv/y) has practically negligible contribution to the average worldwide value. In addition, the average of the total AAIED from radon, radium, and uranium due to ingestion of medicines (2.994 nSv/y) was also found to be much less than the international standards. These data show that the intake of these medicinal plant medicines does not lead to substantial changes of the internal radiation dose.

5 Conclusions

SSNTD of CR-39 is a convenient tool for long-term measurement of alpha particle emitters. Using these detectors, the concentrations of radon, radium, and uranium in 16 pharmaceuticals derived from medicinal plants were studied. The average of the total annual internal effective doses from radon, radium, and uranium due to ingestion of medicines was found to be 2.994 nSv/y. This value is much smaller than the action levels recommended by UNSCEAR and ICRP. As a result, the intake of the current pharmaceuticals does not result in significant changes in the internal radiation dose.

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