MEASUREMENT OF RADON EXHALATION RATE AND ANNUAL EFFECTIVE DOSE FROM MARINE SEDIMENTS, RAS TANURA, SAUDI ARABIA, USING CR-39 DETECTORS

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Abstract. The behavior of radon in marine sediments collected from the beach of Ras Tanura, Arabian Gulf, Saudi Arabia was studied by using radon monitoring system consists of two tightly coupled cup-type plastic containers technique containing SSNTDs of type CR-39 to estimate the radon contribution and to calculate the annual effective dose to inhabitants of the study area. The activity concentration of radon in the sediments was found to be ranging from 13.5 ± 1.4 to 140 ± 11 Bq m^{-3} with an average of 33 ± 2.1 Bq m^{-3}. The calculated values of surface and mass exhalation rates of radon ranged from 5.97 to 61.8 mBq m^{-2} h^{-1} and from 0.11 to 1.47 mBq kg^{-1} h^{-1}, with an averages of 14.6 mBq m^{-2} h^{-1} and 0.30 mBq kg^{-1} h^{-1}, respectively, for the studied samples of marine sediments. The annual effective dose was found to be ranging from 0.34 to 3.54 mSv y^{-1}, with an average of 0.83 mSv y^{-1}. Average value of radon concentrations from marine sediments samples were compared with other literature values.

Key words: Radon, annual effective dose, radon exhalation rates, CR-39.

1. INTRODUCTION

The naturally occurring radioactive materials (NORM) are found throughout the natural environment, in man-made materials such as building materials and fertilizers and in the following crude oil and natural gas operations, which includes long-lived radioactive elements such as ^{238}U, ^{232}Th series and their respective decay daughter (^{226}Ra, ^{228}Ra and ^{222}Rn), as well as ^{40}K. The naturally occurring radioactive materials are relatively and uniformly distributed in the seas and the oceans, which are considered in balance state [1, 2]. However, the discharge into the sea of low level waste from industrial production processes and human consumption activities has become a source of contamination in the marine coastal environment which may be altered the concentration of the natural radionuclides than its physical state [1, 2]. Oil refineries process and shipping of petroleum are produced a huge amount of waste materials which is known as oil refinery sludge. Radium is the most important naturally occurring radionuclides content in sludge. Radon is decay from radium which is found in most of oil, gas fields and oil refinery sludge [2].

Most of the radioactivity deposited on surface sediments is washed by rains and drained through rivers to the oceans. Part of the ground deposited activity is absorbed in the soils and percolates with the underground waters to the seawater. Radionuclides reaching seawater become part of the marine ecosystem (water, sediments, and biota) and may transfer through seawater-sediment-biota interface to human beings [3, 4].

Accumulation of such substances in the marine coastal environment raises many problems concerning safety of biotic life, food chain and ultimately humans. To address these problems, assessment of radioactivity concentration in the marine environment is essential [5]. It is necessary to quantify the distribution of radionuclides in the main marine constituents (sea water, sea sediments and marine organisms) and to assess radiological impacts of the detected radionuclides on human health [5]. Beach sediments are mineral deposits formed through weathering and erosion of either igneous or metamorphic rocks. Among the rock constituent minerals are some natural radionuclides that contribute to ionizing radiation exposure on Earth. The study of the distribution of primordial radionuclides allows the understanding of the radiological implication of these elements due to the radiation exposure of the body and irradiation of lung tissue from inhalation of radon and its daughters [6, 7].

Radon-222 (^{222}Rn) is naturally radioactive gas used to estimate the radioactive hazard due to water, soil and sediment [8]. It is also used to estimate the amount of discharge ground water to the surface water and the amount of radiation in such sediments [9].

Radon-222 produced primarily in sediment by the decay of Ra-226. A fraction of the radon which is produced in sediments will escape to the overlying water column, leaving a deficiency of radon in sediments, so that the activity ratio of radon to radium in less than one. Once radon is in water column, it is mixed vertically and will either decay there or escape to the atmosphere [10].

The study of radon exhalation rates from soils or marine sediments is very important to show the behavior of radon that escapes from soil or sediments into the surrounding air. This may be measured as either due to surface or mass of the sediment sample. The measurement of radon exhalation rate in sediments samples is helpful in determining the health hazard due to radon for the residents and marine life [11–13].

Being aware of the hazardous effects of radon exhalation on human health, it was necessary to conduct measurements of radium content in sediments. Higher values of radium in sediment contribute significantly in the enhancement of environmental radon [14].

The fraction of inhalation due to ^{222}Rn and their products is 1.2 mSv a year, the average worldwide [15].

In the present investigation, radon concentration, mass and surface exhalation rates and the annual effective dose have been estimated for sediment samples collected from different location at Ras Tanura City of the Arabian Gulf (AG), Saudi Arabia. The AG is shallow water body located in a subtropical and hyper arid region [16]. Oil refinery, natural gas and shipping of petroleum which located in Ras Tanura city may generate a lot of health concerns regarding both the population and environment. The information on radon concentration and the annual effective dose of Gulf sediments is very limited in this area, while many studies have been done on radon concentration in other regions of Saudi Arabia [17–24]. However, there is yet lack of published information on radon levels and radon exhalation rates (mass and surface) in sediments for Saudi Arabia. Therefore, the obtained data will provide baseline of radon concentration and the annual effective dose, in coastal sediments of Ras Tanura city, Arabian Gulf, Saudi Arabia for future local and regional studies.

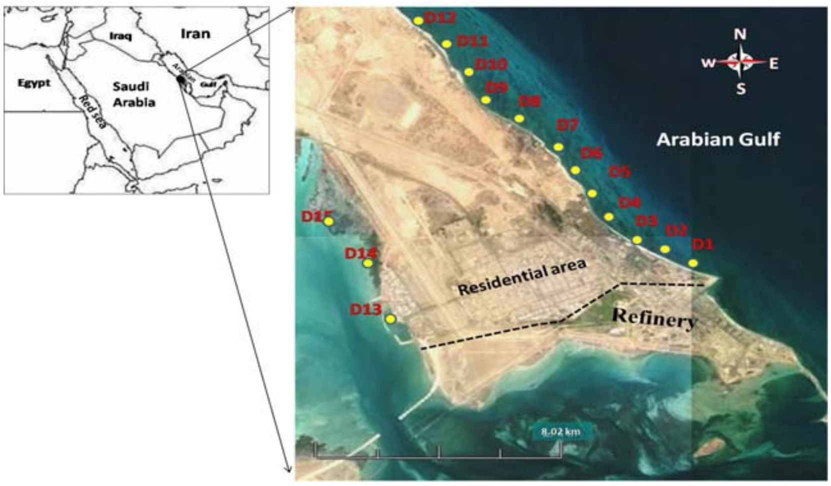

Fig. 1 – Map of the study area (Produced by using Google Earth Software).

2. MATERIALS AND METHODS

2.1. STUDY AREA

Ras Tanura city is located on Arabian Gulf in Eastern Province of Saudi Arabia and its geographical coordinates are 26°38'38" N and 50°9'33" E. Seawater surrounds the city from three sides see Fig. 1. The tide rise, approximately, ranges from 2 to 1.5 meters during a year. Its area is about 290 km² and a population of about 73933 inhabitants. The climate is hot in summer (Absolute maximum 45.6°C in June) with high humidity, it frequently exceeds 85%. Mostly, wind is strong and blowing dust during June and July. The west and south coast of the city are mainly composed of sand, mud, sediments and sludge while the east coast is composed of sand and marine sediments.

Ras Tanura city is one of the most important cities in Saudi Arabia because of the presence of the largest and oldest oil refinery in the Middle East which was began operations in September 1945. Also the study area contains gas plant and two ports for oil export.

2.2. EXPERIMENTAL METHOD FOR RADON MEASUREMENT

In the present study, samples were collected from 15 locations as shown in Fig. 1. One kilogram was collected from each site. Most of the sites near the sludge area along the west coast of the city were not accessible, which was closed by the petroleum companies. Also, the south coast of Ras Tanura city is closed by the oil company. After removing the stones and other particles, all samples were grinded, homogenized and dried in an oven at 70°C for 24 hours. Finally, sediments samples were sieved through 2 mm mesh.

The radon monitoring system consists of two tightly coupled cup-type plastic containers. One of them is a detection chamber and the other is a sample container as shown in Fig. 2. Solid state nuclear track detectors CR-39, 2.5 × 2.5 cm in size, were attached to the bottom of detection chamber in an empty volume of 150 ml. There is a round hole (D = 1.5 cm) in the middle of the lid of the detection chamber, which is covered with a sponge filter. The function of this filter is to reduce the humidity in the detection chamber and to discriminate against thoron ^{220}Rn (half-life is 55.6 s) by hindering its diffusion into the volume of the detection. The two coupled cup-type containers were stored for more than one month to allow radioactive equilibrium to be reached in all samples.

2.3. ETCHING AND SCANNING SYSTEM

After exposure to radon, the detectors were collected from the detection chambers and etched by using 6.25 M NaOH solution (250 grams of solid NaOH made up to one liter of water) at 90 Co in a water bath for 2 hours. After etching, the detectors were removed from the solution and cleaned in distilled water for 15 s, then, transferred to a 2% neutralising solution (by adding 2 parts glacial acetic acid to 98 parts distilled water) for 30 min. Finally, the detectors were removed from the neutralising solution and placed in distilled water for 10 min then placed in a drying cabinet for 20 minutes.

Tracks in CR-39 were automatically counted using an optical microscope (TASLIMAGE scanning and readout system), located at research units, Imam

Abdulrahman Bin Faisal University. A digital CCD camera was attached to the microscope. The camera gives 10 bit images (1024 grey levels). Precise adjustment is not required because the software performs automatic (and continuous) calibrations of the light intensity during scanning. The software automatically displays a live image from the microscope camera. An image of the tracks, due to the alpha particle emitted from radon, was observed directly on a computer screen attached to the microscope.

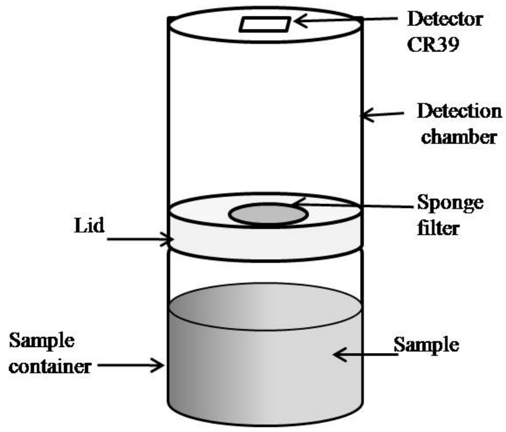

Fig. 2 – The radon monitoring system.

The radon concentration C_{Rn} in detection chamber was calculated using the following equation [25]:

$$C_{Rn}\ (\text{Bq m}^{-3}) = \rho / (K.T) \tag{1}$$

where: ρ is the track density on the detector, T is the exposure time (1440 hours) and K is the calibration factor (tracks cm^{-2} recorded per Bq $\text{m}^{-3}\ \text{h}^{-1}$).

For the purposes of calculating ^{222}Rn concentration and radon exhalation rate, the passive diffusion dosimeter was used; the surface exhalation rate of radon E_S (Bq $\text{m}^{-2}\ \text{h}^{-1}$) is determined by the following formula [12, 26–28]:

$$E_S\ (\text{mBq m}^{-2}\ \text{h}^{-1}) = (V\lambda C/A)/\ T_{eff} \tag{2}$$

where: A is the surface area of sample ($28.26 \times 10^{-4}\ \text{m}^2$).

$$E_M\ (\text{mBq kg}^{-1}\ \text{h}^{-1}) = (V\lambda C/A)/\ T_{eff} \tag{3}$$

where: V is the volume ($150 \times 10^{-6}\ \text{m}^3$), λ is the decay constant of radon gas, M is the mass of samples that ranges between 120 and 170×10^3 kg, C is the calculated integrated radon exposure as measured by SSNTDs of type CR-39 (Bq m^{-3} h) and T_{eff} is the effective exposure time which is defined as:

$$T_{eff} = T - (1/\lambda)(1 - e^{-\lambda T}) \quad (4)$$

The annual effective dose E_{Rn} was calculated using the following equation [15]:

$$E_{Rn}\ (\text{mSv y}^{-1}) = C_{Rn} \times Q_f \times E_f \times D_f \times 24\ \text{h} \times 365 \times 10^{-6} \quad (5)$$

where C_{Rn} is the measured ^{222}Rn concentration (Bq m^{-3}), (D_f) is the conversion factor (effective dose received by adults per unit ^{222}Rn activity per unit of air volume) of 9.0 nSv h^{-1} per Bq m^{-3}, (E_f) is the equilibrium factor that equal to 0.4, (Q_f) is the indoor occupancy factor which equal to 0.8.

3. RESULTS AND DISCUSSION

The activity concentrations of radon and the annual effective doses from sediment samples from Ras Tanura city in the Arabian Gulf, Saudi Arabia are given in Table 1. The activities range and mean values (in brackets) for radon concentration (Bq m^{-3}) and the annual effective dose (mSv y^{-1}) are 13.5 ± 1.4 to 140 ± 11 (33 ± 2.1) Bq m^{-3} and 0.34 to 3.54 (0.83) mSv y^{-1}, respectively.

Table 1

Locations, radon concentrations and annual effective doses in sediments samples

Sample code	Latitude	Longitude	C_{Rn} (Bq m^{-3})	E_{Rn} (mSv y^{-1})
D1	26°42′36.7″ N	50°5′47.3″ E	15.6 ± 1.7	0.39
D2	26°42′49.8″ N	50°5′19.9″ E	21.1 ± 2.2	0.53
D3	26°43′9.50″ N	50°4′48.3″ E	16.7 ± 1.8	0.42
D4	26°43′38.6″ N	50°4′22.4″ E	18.1 ± 1.9	0.46
D5	26°44′6.00″ N	50°4′6.40″ E	38.3 ± 3.4	0.97
D6	26°44′27.4″ N	50°3′53.7″ E	13.5 ± 1.4	0.34
D7	26°44′49.3″ N	50°3′3.20″ E	18.1 ± 1.9	0.46
D8	26°45′7.00″ N	50°3′7.00″ E	15.9 ± 1.7	0.40
D9	26°45′24.3″ N	50°2′43.0″ E	37.0 ± 3.3	0.93
D10	26°45′44.5″ N	50°2′29.8″ E	19.9 ± 1.8	0.50
D11	26°46′12.3″ N	50°2′14.2″ E	17.8 ± 1.4	0.45
D12	26°46′44.3″ N	50°1′42.2″ E	37.6 ± 3.0	0.95
D13	26°43′4.90″ N	50°1′10.1″ E	20.6 ± 1.8	0.52
D14	26°43′7.80″ N	50°0′33.3″ E	65.3 ± 3.9	1.65
D15	26°43′40.7″ N	50°0′32.4″ E	140 ± 11	3.54
Min value			13.5 ± 1.4	0.34
Max value			140 ± 11	3.54
Average value			33 ± 2.1	0.83

The maximum activity concentration of radon and annual effective dose of 140 ± 11 Bq m^{-3} and 3.54 mSv y^{-1}, respectively, were observed for sediment sample

D15 which was collected from the west shoreline of the city where the sludge area is situated. These higher values were interpreted to that the location of the picked sample is observed to be adjacent and very close to the sludge area which leaked to the environment due to the oil refinery process and shipping of petroleum. The naturally occurring radionuclides waste (NORM) is concentrated and accumulated in the environment in the form of sludge and scale [29]. Radon decays from radium which may content in the naturally occurring radionuclides waste, NORM. Further, the location of Sample D15 was observed to be adjacent to the sedimentary rocks that have undergone great stress, change and fracturing. The lowest radon concentration value and annual effective dose for the studied marine sediments samples are found to be 13.5 ± 1.4 Bq m^{-3} and 0.34 mSv y^{-1}, respectively. These lower values were recorded for the marine sediment sample for site D6, this site is located on the east shoreline within a flat area which are near to sandy area and far from the sludge area. Low concentrations value of radon due to radium contents in the earth's crust and their concentrations depend mainly on the local geology geographical conditions which appear at different levels in each region in the world [15].

Figures 3 and 4 show the variations of radon concentrations and annual effective doses at different sampling locations. The distribution of the values of radon concentration and annual effective dose show that the values on the west coast of the city tend to rise, where sludge area located, as shown at sites D14 and D15. The wide variations of the activity concentration values are due to their presence in the marine environment and their physical, chemical and geo-chemical properties [30, 31]. On the other hand, variation among the activity concentrations of radon of the collected samples from different locations may be due to the continuous wave action, where the high tide may lead to the deposition of radionuclides in sediments such as radium and uranium along the shoreline.

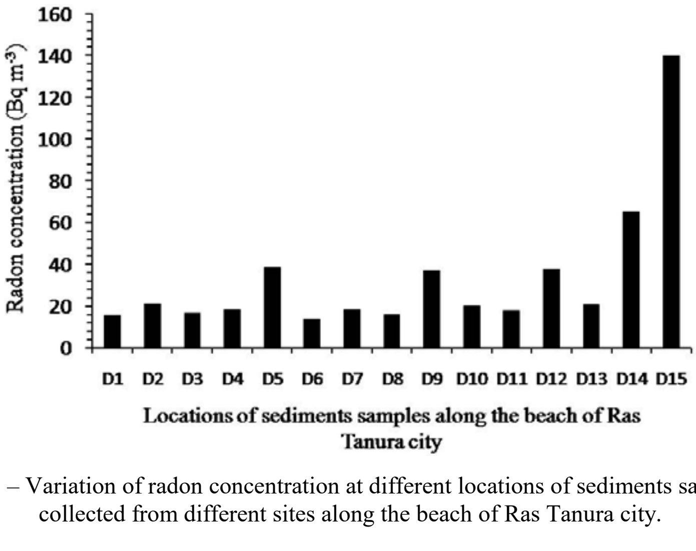

Fig. 3 – Variation of radon concentration at different locations of sediments samples collected from different sites along the beach of Ras Tanura city.

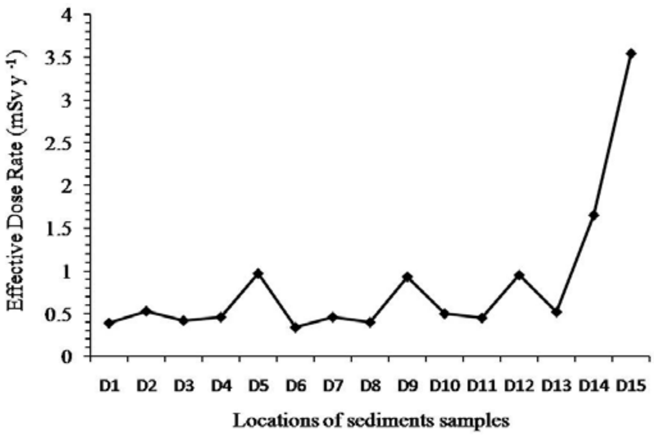

Fig. 4 – Variation of the annual effective dose with the locations of sediments samples collected from different sites along the beach of Ras Tanura city.

The range and mean values (in brackets) for the surface and mass radon exhalation rates, E_s and E_M, obtained in these measurements are shown in Table 2. The results show that the surface and mass exhalation rates of radon varies from 5.97 to 61.8 (14.6) mBq m^{-2} h^{-1} and from 0.11 to 1.47 (0.30) mBq kg^{-1} h^{-1}, respectively. The highest values of surface and mass exhalation rates of radon of 61.8 mBq m^{-2} h^{-1} and 1.47 mBq kg^{-1} h^{-1}, respectively.

Table 2

Surface and mass exhalation rates of radon gas in sediments

Sample code	E_s (mBq m^{-2} h^{-1})	E_M (mBq kg^{-1} h^{-1})
D1	6.89	0.13
D2	9.28	0.18
D3	7.34	0.14
D4	7.96	0.15
D5	16.9	0.32
D6	5.97	0.11
D7	7.96	0.15
D8	6.99	0.13
D9	16.3	0.31
D10	8.77	0.17
D11	7.85	0.15
D12	16.6	0.31
D13	9.08	0.17
D14	28.8	0.54
D15	61.8	1.47
Min value	5.97	0.11
Max value	61.8	1.47
Average value	14.6	0.30

It is clear to see that the surface and mass exhalation rates vary from point to point as shown in Figs. 5 and 6. Similarly, the highest value is noted for sediment sample D15 which collected from the site near sludge area. The variations of radon activity, E_s and E_m depend on the sludge that accumulated along the shoreline. Sludge area could increase the exhalation rate of radon in sediment due to the radium contents. However, these variations of radon activity, E_s and E_m may also depend on local characteristics of sediment for the studied area. This explanation can be clearly seen in Figs. 5 and 6, where the values of the mass and surface exhalation rates increase at sites D14 and D15, where these two sites are adjacent to the sludge area and located on the west coast of the city.

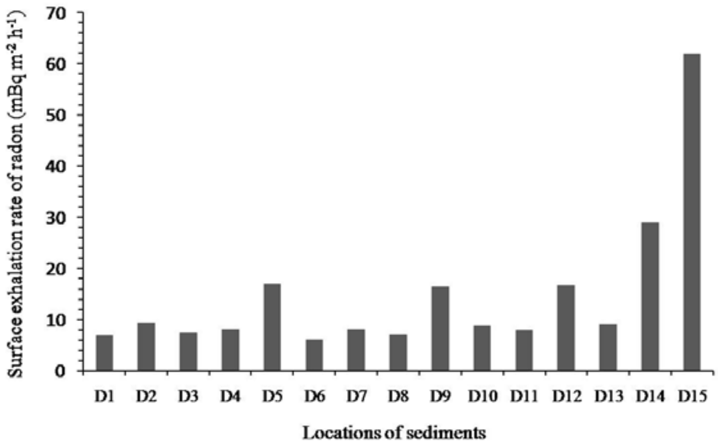

Fig. 5 – Surface exhalation rate of radon *vs.* locations of sediments samples collected from different sites along the beach of Ras Tanura city.

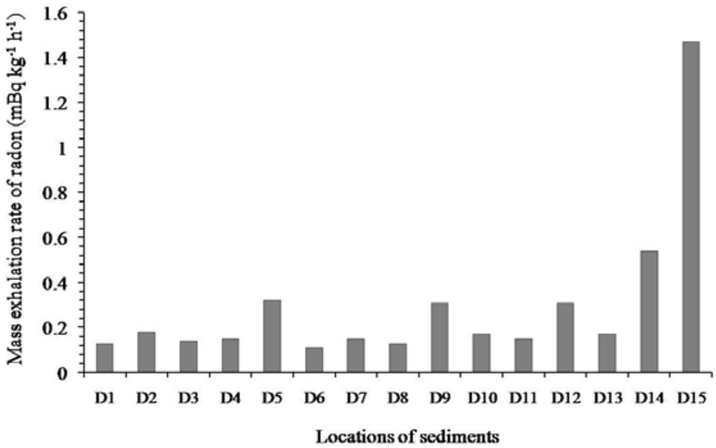

Fig. 6 – Mass exhalation rate of radon *vs.* locations of sediments samples collected from different sites along the beach of Ras Tanura city.

It is very important to notice that, this study is constitute the data baseline for the surface and mass exhalation rates in addition to the annual effective dose due to radon from marine sediment samples.

Table 3 compares the radon activity concentrations in beach sediment samples with other studies in different beaches of the world [32–39]. The average of radon concentration in sediment of our study was lower than the value from west of Saudi Arabia where the volcanic and granite rocks exist, while it is slightly higher than that value from Thailand. Generally, the obtained value of radon is lower than the other studies from Czech Republic, Iraq, Jordan, China, British and India. On the other hand, it is suitable to conclude that the recorded values are seen to be lower than the hazardous values when compared with other average values from other national and worldwide works.

Table 3

Comparison of radon concentration level in sediment samples with local and worldwide results

Country	C_{Rn} (Bq m^{-3})	Reference
Ras Tanura – Arabian Gulf – Saudi Arabia (East)	33 ± 2.1	Present study
Aqaba – Saudi Arabia (West)	173	[32]
Bohemian Massif – Czech Republic	162.6	[33]
Khor-Abdulla – Iraq	304.3 ± 79.4	[34]
Thailand	4.5 ± 0.6	[35]
Jordan	918 ± 214	[36]
China	37.64	[37]
Dee River – UK	33–233	[38]
Hemavathi River – India	334.90	[39]

4. CONCLUSION

Knowledge about the occurrence and concentration of radon in environmental sediment samples is essential for monitoring their background levels and their health effects for population in the coastal areas. Provide insight into changes in their concentrations that may result from anthropogenic inputs. This knowledge can be useful in understanding the effects of radon levels as in the environment and also for the estimation of radiological hazards to human health. Baseline data of the present study provides a platform for measuring the radon concentration levels and the annual effective dose in the sediment samples collected from the beach of Ras Tanura city, Arabia Gulf, Saudi Arabia. The recorded results of our study is representing the activity concentrations of radon in the sediments that range between 13.5 ± 1.4 ~ 140 ± 11 Bq m^{-3} with an average of 33 ± 2.1 Bq m^{-3}, the values of surface and mass exhalation rates ranged from 5.97 to 61.8 mBq m^{-2} h^{-1} and from 0.11 to 1.47 mBq kg^{-1} h^{-1} with an averages of 14.6 mBq m^{-2} h^{-1} and 0.30 mBq kg^{-1} h^{-1}, respectively. The average value of annual effective dose was found to be 0.83 mSv y^{-1}. The results for radon concentrations from the sediments of Ras Tanura beach were compared with other literature values and seems to be acceptable and not concerning a health hazard for the inhabitant of the town.

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