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Research Article
Characterization of Radon Concentration and Annual Effective Dose of Soil Surrounding a Refinery Area, Ras Tanura, Saudi Arabia

[1]Fatimh Alshahri, [2]Atef El-Taher and [3,4]Abd Elmoniem Ahmed Elzain

[1]Department of Physics, College of Science, Imam Abdulrahman Bin Faisal University, P.O. Box 1982, 31441 Dammam, Saudi Arabia
[2]Physics Department, Faculty of Science, Al-Azhar University, 71452 Assuit, Egypt
[3]Department of Physics, Kassala University, Kassala, P.O. Box 266, Sudan
[4]Department of Physics, College of Science and Arts in Oklat Al Skoor, Qassim University, P.O. Box 111, Buraydah, Saudi Arabia

Abstract
Background and Objective: Oil refineries process and shipping of petroleum contaminate the environment due to leakage of waste materials containing naturally occurring radionuclides. Radon decays from radium which may content in the waste. Therefore, the present work dealt with measuring soil gas radon concentration, surface and mass exhalation rates from soil samples surrounding a refinery area in Ras Tanura city, Saudi Arabia. **Materials and Methods:** The measurements were carried out using CR-39 detectors. After etching process, all detectors were automatically counted using scanning and readout system and theoretical calculations were done to find the radon concentration, radon surface and mass exhalation rates and Dose estimation. **Results:** The soil gas radon concentration ranged from 6.02$\pm$0.8 to 927$\pm$101 Bq m^{-3} with an average value of 120$\pm$14 Bq m^{-3}. The surface and mass exhalation rates were found to be ranging from 2.65-409 mBq m^{-2} h and from 0.04-8.31 mBq kg^{-1} h with an average of 53.4 mBq m^{-2} h and 1.08 mBq kg^{-1} h, respectively and the effective dose rate ranged from 0.15-23.4 mSv/y with an average of 3.02 mSv/y. A good positive correlation coefficients were observed between radon concentration and surface and mass radon exhalation rate of soil samples. The obtained results compared with other findings from local and worldwide locations. **Conclusion:** The obtained results were found to be within the recommended limits. This study could be useful as a baseline data for monitoring and evaluation radon exposure in soil around the residential area nearby refinery area in Arabian Gulf region.

Key words: Radon, surface and mass exhalation rates, annual effective dose rate, CR- 39, g-ray emissions

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Corresponding Author: Fatimh Alshahri, Department of Physics, College of Science, Imam Abdulrahman Bin Faisal University, P.O. Box 1982, 31441 Dammam, Saudi Arabia Tel: +966-133337205

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Men's exposure pathways to natural radiation have two distinct components, the external component due to the r-ray emissions and the internal one partly due to radon and its decay products which are a-particle emitters[1]. In fact, ^{222}Rn and its progeny are responsible for about two third of the exposure from natural sources[1]. Radioactivity enters the human body mainly by inhalation of radon and thoron and their decay products[2] and also by ingestion of primordial radionuclides and their progeny, such as ^{40}K, ^{238}U and ^{232}Th series radionuclides[3].

Soils have different concentrations of radionuclides depending on their formation from the parent rocks, geographical location and by geochemical processes[4]. As an inert gas, radon freely diffuses through the soils and reaches the atmosphere where it could migrate into structure of dwellings to pose a health hazard. Inhalation of radon (^{222}Rn) and its short-lived daughters, namely ^{218}Po and ^{214}Po are associated with the risk of lung cancer, if radium content was sufficiently high and ^{222}Rn was used to estimate the radioactive hazard due to soil and sediment[5].

Radon gas formed from the decay process of uranium, with several intermediate progenies. The ^{222}Rn produced primarily in soils by the decay of ^{226}Ra. A fraction of the radon which is produced in the soil 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[5]. Radon concentrations in soil gas within a few meters of the surface of the ground are clearly important in determining radon rates of entry into pore spaces and subsequently into the atmosphere and it's depend on the radium concentration in the bedrock and on the permeability of the soil[1,6-8]. The radon surface and mass exhalation rates are depending upon a number of parameters that behave in a stochastic and independent fashion, such as the radioactive disintegration of ^{226}Ra to produce radon, the direction of recoil of radon in the grain, the interstitial soil moisture condition in the vicinity of the ejected radon atom and its diffusion in the pore space[9].

Radioactive materials which occur naturally and expose people to radiation occur widely. The NORM is an acronym for Naturally Occurring Radioactive Material, which includes long-lived radioactive elements, such as uranium and thorium and their daughter products[10], oil and gas production and processing operations sometimes cause NORM to accumulate at elevated concentrations in by-product waste streams[11,12]. In addition, radon gas, a radium daughter, may be found in produced natural gas. In gas processing activities, NORM generally occurs as radon gas in the natural gas stream[13].

The aim of the present work was to evaluate the distribution of soil gas radon concentration, surface and mass exhalation rates of radon and annual effective dose for soil samples collected from Ras Tanura, Arabian Gulf- Saudi Arabia. This study is the first investigation in this area which could contribute to establish a database to estimate the increase of radon concentration nearby the oldest refinery in Saudi Arabia. Moreover, this study will provide scientific information that may be useful for authorities in the implementation of radiation protection standards for the general public.

MATERIALS AND METHODS

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. The tide rise, approximately, ranged from 2-1.5 m during a year. This area is about 290 km^2 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. 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 2 ports to oil ship. West and South of the city, coast is mainly composed of sand, mud, sediments and sludge while the East coast is composed of sand and marine sediments.

Experimental method for radon measurement: Sampling was carried out from 34 locations taking place along Ras Tanura city (Fig. 1). One kilogram was collected from each site. Some points near the sludge area (South of Ras Tanura city) were not accessible, which were closed by the oil company. All samples were ground, homogenized and dried in an oven at 70°C for 24 h. Finally, soil samples were sieved through 2 mm mesh.

The radon monitoring system consists of two tightly coupled cup-type plastic containers[14]. One of them was a detection chamber and the other was sample container (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 was a round hole (D = 1.5 cm) in the middle of the lid of the detection chamber, which was covered with a sponge filter. The function of this filter was 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

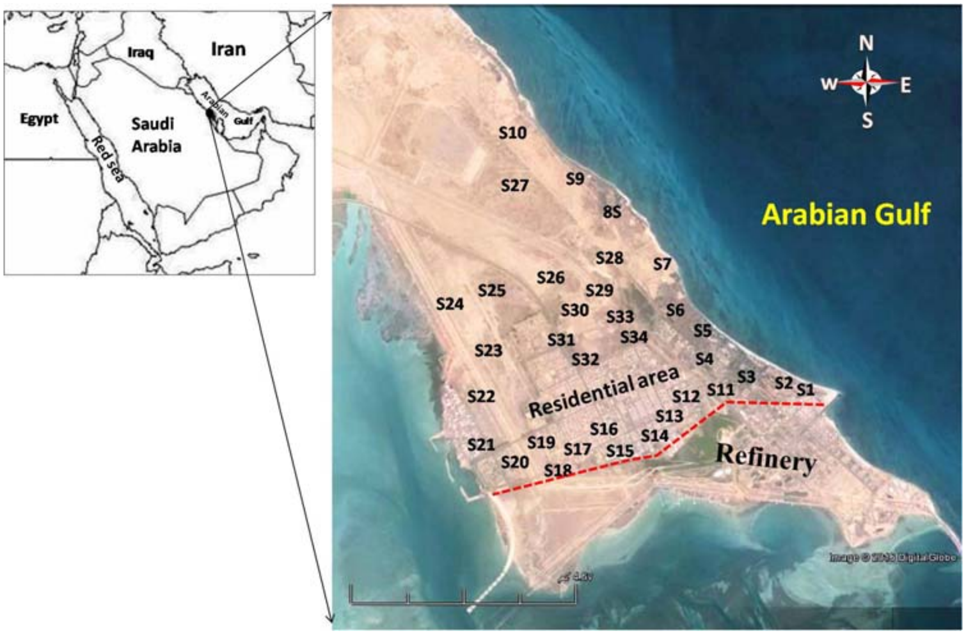

Fig. 1: Map of the study area near a refinery, Ras Tanura city, Arabian Gulf, Saudi Arabia (Produced by using Google Earth Software)

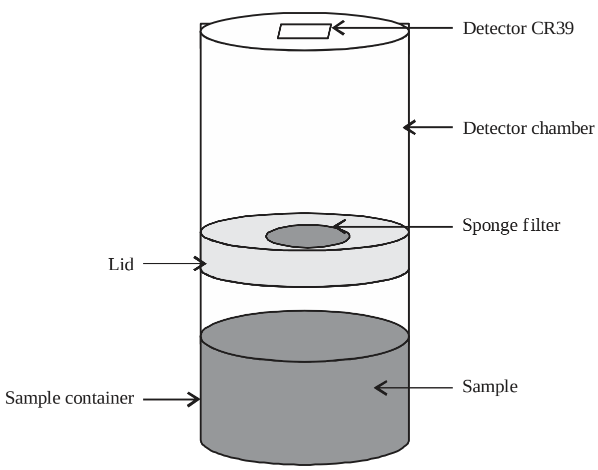

Fig. 2: Radon monitoring system

two coupled cup-type containers were stored for more than one month to allow radioactive equilibrium to be reached in all samples.

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 g of solid NaOH made up to 1 L of water) at 90°C in a water bath for 2 h. After etching, the detectors were removed from the solution and cleaned in distilled water for 15 sec, then, transferred to a 2% neutralizing solution (by adding 2 parts glacial acetic acid to 98 parts distilled water) for 30 min. Finally, the detectors were removed from the neutralizing solution and placed in distilled water for 10 min then placed in a drying cabinet for 20 min. Tracks in CR-39 were automatically counted using an optical microscope (TASLIMAGE scanning and readout system), located at research units laboratory, 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 was 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.

Theoretical calculations

Radon concentration: The radon concentration C_{Rn} in detection chamber was calculated using the following equation[15]:

$$C_{Rn} = \frac{\rho}{T_{\eta}}\left(Bq\ m^{-3}\right) \quad (1)$$

Where:

ρ = Track density (tracks per cm^2)

T = Exposure time and η is the calibration factor

Radon surface and mass exhalation rates: In order to measure radon concentration and its exhalation rate, using the sealed can technique, the radon exhalation rate in terms of area E_s (mBq m^{-2} h) is calculated by using Eq. 2, as[16,17]:

$$E_s = \frac{\lambda VC}{A\left[T + \lambda^{-1}\left(\exp\left(-\lambda T\right) - 1\right)\right]} \quad (2)$$

where, C is the integrated radon exposure (Bq m^{-3} h), l is the radon decay constant, V is effective volume of detection chamber and A is the surface area of the sample (m^2).

Moreover, the radon exhalation rate in terms of mass E_M (mBq kg^{-1} h) is determined by Eq. 3[16,17]:

$$E_M = \frac{\lambda VC}{M\left[T + \lambda^{-1}\left(\exp\left(-\lambda T\right) - 1\right)\right]} \quad (3)$$

where, M is the mass of sample (kg).

Dose estimation: The calculation of the annual effective dose E_{eff} was done by converting the average radon concentration C_{Rn} into the following equation[18].

$$E_{eff}\ (mSv/y) = C_{Rn} \times Q_f \times E_f \times D_f \times 8760 \quad (4)$$

Where:

C_{Rn} = Measured ^{222}Rn concentration (Bq m^{-3})
D_f = Conversion factor of 9.0 ' 10^{-6} mSv/h per Bq m^{-3}
E_f = Equilibrium factor that equal to 0.4
Q_f = Indoor occupancy factor which equal to 0.8 and 8760 is the number of hours per year

RESULTS AND DISCUSSION

The results of radon concentration from soil samples from Ras Tanura, Arabian Gulf, Saudi Arabia are presented in Table 1 and Fig. 3. The measurements carried out on soil gas radon concentration reveal a variation from 6.02±0.8 Bq m^{-3} in location S2 to 927±101 Bq m^{-3} in location S18, the overall average was found to be 120±14 Bq m^{-3}. The lower value of soil gas radon concentration was recorded at location S2 may be attributed to the type of soil which was observed to be fine dense sandy soil beside that it was observed to be with low porosity, soil texture and composition was very important in determining the radon concentration value[19,20]. The soils with low porosity are play an important role in decreasing the radon concentration values due to the lower diffusion coefficient[21,22]. It may also be due to the presence of low existence of uranium mineralization and content in the soil of this area. In contrast, it was recorded that higher radon reported for areas due to the presence of high existence of uranium mineralization[23].

The higher value of soil gas radon concentration was recorded at Location S18, this sample was observed to be adjacent and very close to sludge area and noticed to be mixed with a large amounts of suspended materials which was thought to be re-sedimented, this may be attributed to

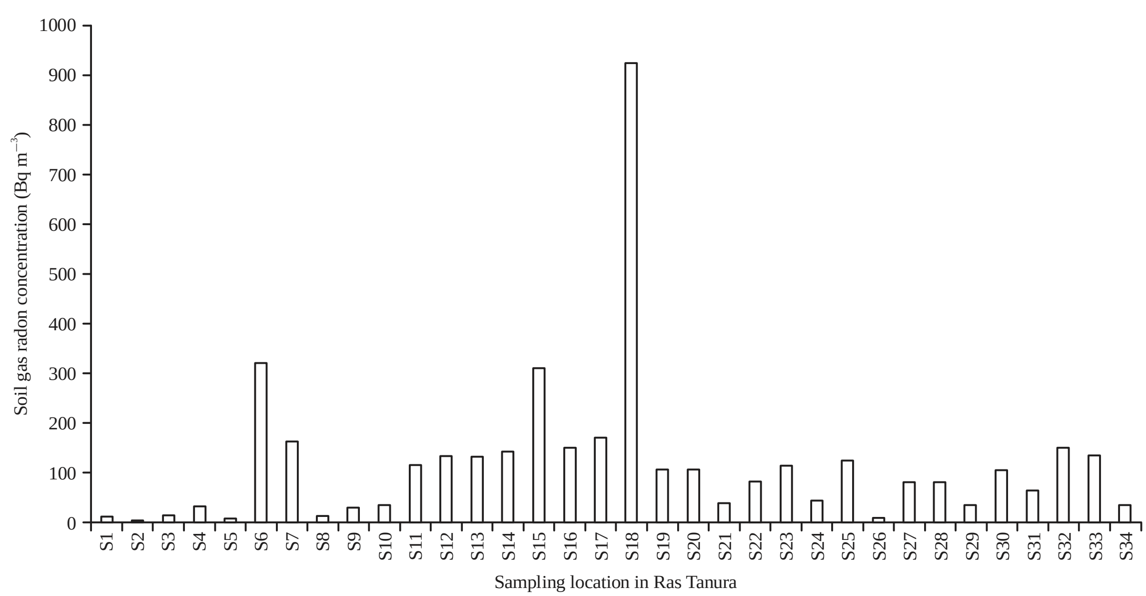

Fig. 3: Soil gas radon concentration (Bq m^{-3}) vs. sampling location in Ras Tanura

Table 1: Locations, radon concentrations, surface and mass radon exhalation and effective dose rates for soil samples from Ras Tanura, Saudi Arabia

	Location					
Sample codes	Latitude	Longitude	CR_n (Bqm^3)	E_s (mBq m^{-2} h)	E_M (mBq kg^{-1} h)	E_{eff} (mSv/y)
S1	26°42'29.1"N	50°5'46.7"E	13.70±2.1	6.02	0.10	0.34
S2	26°42'45.9"N	50°5'20.1"E	6.02±0.8	2.65	0.04	0.15
S3	46°42'59.1"N	50°4'57.8"E	14.20±1.9	6.27	0.09	0.36
S4	26°43'15.4"N	50°4'35.2"E	33.70±4.7	14.8	0.25	0.85
S5	26°42'44.3"N	50°4'51.7"E	9.03±1.4	3.98	0.07	0.23
S6	26°43'15.7"N	50°4'18.0"E	322.00±38	142	2.43	8.11
S7	26°43'26.4"N	50°3'57.1"E	165.00±21	72.8	1.10	4.17
S8	26°44'25.1"N	50°3'45.2"E	15.70±2.2	6.94	0.11	0.40
S9	26°45'35.5"N	50°2'15.6"E	31.10±4.0	13.7	0.25	0.79
S10	26°45'58.6"N	50°1'59.3"E	36.30±5.1	16.0	0.25	0.92
S11	26°42'18.7"N	50°4'39.5"E	117.00±15	51.5	1.05	2.95
S12	26°42'15.8"N	50°4'35.7"E	137.00±19	60.5	1.01	3.46
S13	26°42'5.20"N	50°4'20.6"E	132.00±17	58.2	1.21	3.33
S14	26°41'52.2"N	50°4'9.30"E	144.00±20	63.6	1.26	3.64
S15	26°41'48.4"N	50°3'53.7"E	312.00±37	138	3.60	7.88
S16	26°41'44.2"N	50°3'27.8"E	151.00±21	66.6	1.65	3.81
S17	26°41'44.6"N	50°3'8.50"E	172.00±24	76.0	2.01	4.35
S18	26°41'42.1"N	50°2'21.8"E	927.00±101	409	8.31	23.4
S19	26°41'50.1"N	50°2'21.8"E	107.00±14	47.3	0.88	2.71
S20	26°41'49.7"N	50°1'58.7"E	108.00±15	47.4	0.90	2.71
S21	26°42'14.6"N	50°1'51.1"E	38.90±5.8	17.1	0.35	0.98
S22	26°42'37.3"N	50°1'38.9"E	82.50±13	36.4	0.56	2.08
S23	26°43'4.70"N	50°1'27.1"E	115.00±16	50.7	1.05	2.90
S24	26°43'32.9"N	50°1'9.70"E	46.30±6.9	20.4	0.35	1.17
S25	26°43'41.7"N	50°1'22.4"E	127.00±18	56.0	1.16	3.21
S26	26°45'7.70"N	50°1'58.6"E	9.61±1.4	4.23	0.09	0.24
S27	26°45'31.3"N	50°1'28.9"E	80.60±12	35.5	0.72	2.03
S28	26°43'57.6"N	50°3'16.4"E	80.60±13	16.8	0.34	2.03
S29	26°43'34.4"N	50°3'11.7"E	38.10±5.7	46.7	1.10	0.96
S30	26°43'22.1"N	50°2'52.8"E	106.00±14	28.9	0.62	2.67
S31	26°43'4.90"N	50°3'0.40"E	65.60±9.8	67.2	1.26	1.66
S32	26°43'13.3"N	50°3'23.0"E	152.00±21	60.0	1.10	3.85
S33	26°43'4.00"N	50°3'28.2"E	136.00±18	16.0	0.32	3.43
S34	26°42'52.2"N	50°3'6.60"E	36.20±5.4	55.7	1.10	0.91
Min			6.02±0.8	2.65	0.04	0.15
Max			927.00±101	409	8.31	23.4
Average			120.00±14	53.4	1.08	3.02
Std. deviation			162	71.2	1.48	4.02

the probability of high content of effective radium in the sampling location[14]. The observed concentration of radon in soil of this area was controlled by the presence of petroleum production and associated different thrust that help in easy escape of gases from the deeper part of the crust. It has been also noticed that the coarse sediment cover on the study area may deplete the radon contents substantially through the earth crust of the study area. The lithological controls on radon content of the overlying soil seem to be less prevalent compared to structural and tectonic discontinuities[24]. The potential radon level in the soil could be evaluated from the measured radium content[25]. It can be seen from the results that the soil gas radon concentration varies significantly among samples from the sampling location to another throughout the study area. The differences may due to the lightly variations of radium and uranium contents of some of the samples, which result in higher exhalation rates.

Surface exhalation rate (mBq m^{-2} h) and mass exhalation rate (mBq kg^{-1} h) of radon for the studied samples are presented in Table 1. Surface exhalation rate ranged from 2.65-409 mBq m^{-2} h in locations S2 and S18, respectively, the average value was 53.4 Bq m^{-2} h. Mass exhalation rate ranged from 0.04-8.31 mBq kg^{-1} h, in locations S2 and S18, respectively, the average value was 1.08 mBq kg^{-1} h. However, average value for surface exhalation rate of radon was slightly lower than the worldwide average of 57.6 Bq m^{-2} h (0.016 Bq m^{-2} sec)[18].

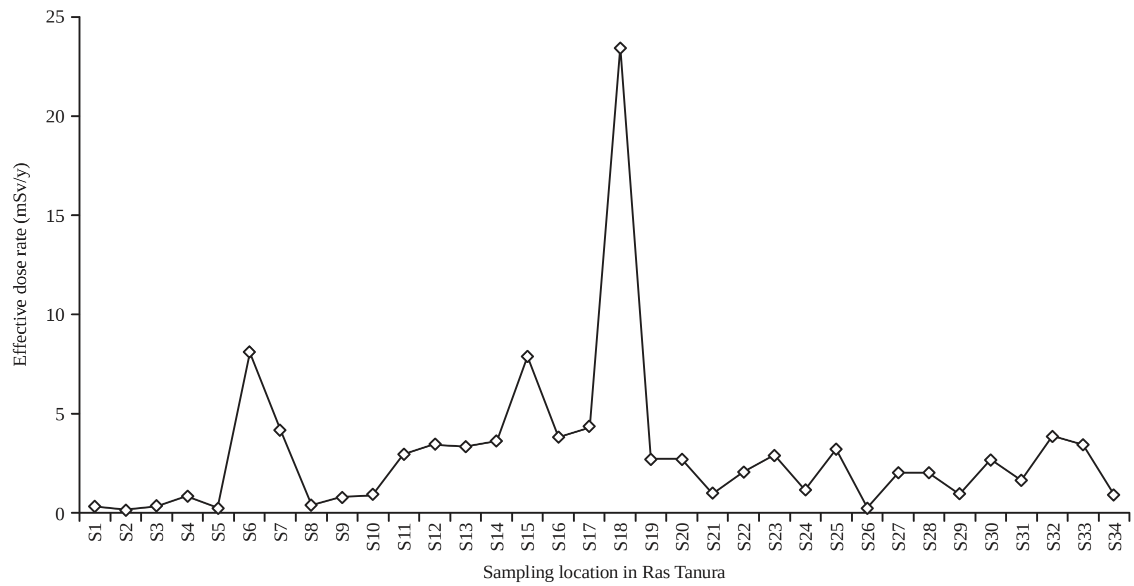

Fig. 4: Annual effective Dose Rate (mSv/y) vs. sampling location in Ras Tanura

It was clear that the results of surface exhalation rate (mBq m^{-2} h) and mass exhalation rates of radon (mBq kg^{-1} h) are strongly related to the radon concentration value of the study material[17], it seems that some of the most important parameters that determine the intensity of radon exhalation from the soil are radium content of the soil[26,27]. It had been expected that exhalation rates of radon should depend on the uranium and radium concentrations in such studied samples and also depends on many other factors, such as permeability, porosity, density, texture and grain size[28]. The radon exhalation rate study was important for understanding the relative contribution of the material to the total radon concentration found in the soil samples and helpful to study radon health hazard[29,30].

Table 1 and Fig. 4 summarized the results of the annual effective dose rate (mSv/y), due to the soil gas radon concentration from the study area. The annual effective dose rate (mSv/y) was found to be ranging from 0.15 mSv/y in Location S2 to 23.4 mSv/y in Location S18, with an average value of 3.02 mSv/y. The higher value of effective dose rate was recorded at Location S18 this may be due to the sample site, where it was observed near to the location of sludge area, which may contain elevated levels of radium[13]. On the other hand, the sample was noticed to be mixed with large amounts of suspended materials which was thought to be re-sedimented, this may attributed to the reason of high content of effective radium in the sampling location[14].

The lower values of effective dose rates from this study were collected from some locations which are seen to be an open areas. The lower recorded values may be due to the wash out of radioactive minerals which contributes to the total effective dose rate in these sampling positions due to the probable rain fall. From present results it was clear that the average annual effective dose was about three times the acceptable value of annual effective dose 1 mSv/y for the public as recommended by the International Commission on Radiological Protection (ICRP) for the individual members of the public[31] and was higher than the world average value (0.07 mSv/y)[18]. The values of annual effective dose do not exceed limits recommended by the european commission (EC)[32]. For the purposes of nuclear safety the results of this study can lead us to conclude that the health risks due to radon for the soil of the study area were very low. Hence, this study results clearly showed that the soil in the study area under investigation is safe as far as the radiological health hazards of radon are concerned.

Figure 5 and 6 summarize the correlation between radon concentration with surface and mass exhalation rates from soil samples. A graph has been plotted between radon concentration and surface and mass radon exhalation rates. A good correlation has been observed between radon concentration and surface exhalation rates with a correlation coefficient of R = 0.961 (Fig. 5). Similarly, the correlation coefficient between radon concentration and mass radon exhalation rates was taking the value of R = 0.942 as shown in Fig. 6. The regression was found linear and positive with a correlation coefficient. The linear type of correlation may be attributed for that the values of exhalation rate depend on

Table 2: Comparison between the results of radon concentration, surface and mass exhalation rates from Ras Tanura soil with other results from worldwide

Country	C_{Rn} (Bq m^{-3})	E_s (mBq m^{-2} h)	E_M (mBq kg^{-2} h)
Ras Tanura, Saudi Arabia (East) (present study)	120	53.4	1.08
Saudi Arabia, Jeddah (West)[35]	36	(4.58-8.40) ‘ 10^3	135-251
Saudi Arabia, Najran (South)[37]	990-1400	-	-
Benghazi, Libya[28]	220.3	216.5	8.2
Nigeria[13]	-	2.60-6.25	-
Kenya[38]	35±14	-	-
India[39]	330.5±30.4	119.1±11.1	4.6±0.4
Iraq[40]	304.3	0.267	6.87
Khajjiar, India[41]	-	502.12-1162.64	15.16-35.11
Kosovo[42]	0.295-32	-	-
Slovenia[36]	900-32900	1584-60336 (1.1-41.9 mBq m^{-2} sec)	-
Niška Banja town, Serbia[43]	33765	-	-
Southern Punjab, Pakistan[44]	34-260	230-288	-
Sudan, Rabak[22]	8.20' 10^3	7.2	145
Cameron[25]	(6.7-10.8)10^3	-	-
Turkey[45]	98-8594	-	

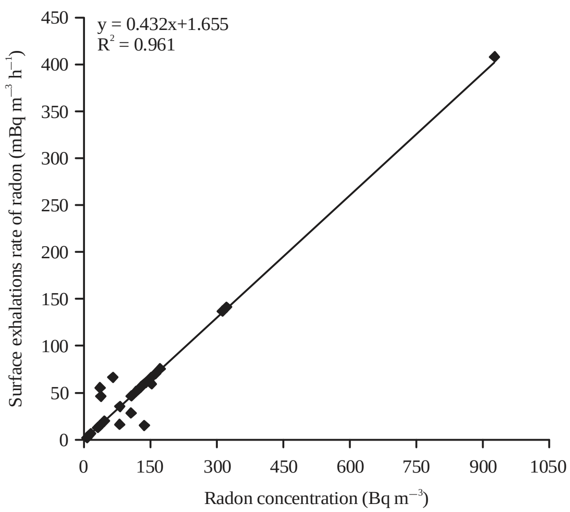

Fig. 5: Surface exhalation rate of radon (mBq m^{-2} h) vs. radon concentration (Bq m^{-3}) in Ras Tanura

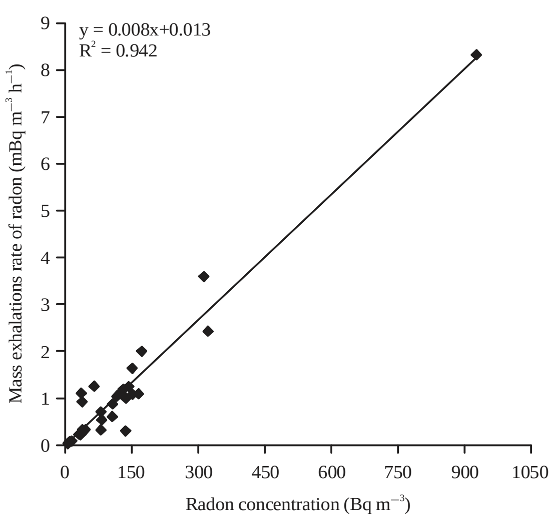

Fig. 6: Mass exhalation rate of radon (mBq kg^{-1} h) vs. radon concentration (Bq m^{-3}) in Ras Tanura

radon concentration since the volume of the cup, the area of the sample and decay constant of radon are the same for all samples. In addition to that the positive correlation predicts that the samples collected in this region are geochemically coherent.

Table 2 shows comparison between radon concentration, surface and mass exhalation rates measured in soil samples from different locations with reported data by other investigators from literature. In general, the radon concentrations of soil samples from Ras Tanura city were lower than most other values of soil gas concentration reported in Table 2. Furthermore, the obtained values of soil gas radon concentration were found to be lower than the range characteristic of deep soil radon concentration (10^3 to 40×10^3 Bq m^{-3})[33]. Besides, the average of radon concentration was lower than the value recorded in soil by the international commission on radiological protection[34].

The averages of surface and mass exhalation rates are lower than recorded values from other studies, with the exception of the values from Sudan, Nigeria and Iraq for surface exhalation rate. By comparing the result with local studies, it is clear that this study of the values of radon exhalation rates in soil are quite low as compared with that reported in the soil of western Saudi Arabia[35]. This variation may due to the fact that radon exhalation rate depends on local characteristics such as thickness of soil, grain size, radium content, porosity of soil and other geophysical and geochemical parameters[20,36].

CONCLUSION

Radon concentration, surface and mass exhalation rates from soil samples nearby a refinery in Ras Tanura city, Saudi Arabia, were measured using the can technique containing CR-39. The overall computed average of soil gas radon concentration for all sampling location is lower than the range characteristic of deep soil radon concentration. The average value of the effective dose rates was slightly larger than the acceptable value of annual effective dose for the public as computed by UNSCEAR and ICRP. However, the surface exhalation rates are lower than the worldwide average and are found to be strongly and linearly correlated and related to the radon concentrations values of the soil samples. Finally, the soil in this region is safe and there is no radiological health hazard to the population due to radon exposure.

SIGNIFICANCE STATEMENT

This study discovers the radon concentrations in soil surrounding residential area nearby oil refinery within the allowed limit. This study will help the researchers to evaluate the radioactive hazard to the community residents. Further, the results will provide the researchers with the environmental baseline information to evaluate the increase of radon gas in the study area.

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