Measurement of exposure to radionuclides (^{40}K, ^{226}Ra, and ^{232}Th) in the oil and gas drilling industry

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ABSTRACT

Background: Naturally occurring radioactivity is a root cause of human exposure to harmful radiation. The occupational exposure hazard due to natural radionuclides occurring in drilling wastes is especially important in this regard. In this study the concentration of radionuclides namely ^{232}Th, ^{226}Ra and ^{40}K were assessed in soil samples that were taken from various oil drilling areas. ***Materials and Method:*** 10 samples were collected and sealed for two months to ensure the secular equilibrium between ^{226}Ra and ^{232}Th, and their respective radioactive progenies. The concentration of radionuclides in samples was measured by gamma spectroscopy. Descriptive and analytical statistics were used in order to analyze the data. ***Results:*** the results showed the average absorbed dose rates (D), annual effective dose (AED), Radium equivalent activity (Raeq) and various hazard indexes(Hex, Hin and Iγ) for samples were 38.22 nGy/h, 0.046 mSv/y, 81.032 Bq/Kg, 0.21, 0.31 and 0.59, respectively. ***Conclusions:*** The mean activity concentrations were lower than the world mean values, according to the radiation protection criteria that identified by UNSCEAR.

Keywords: *Exposure, natural radioactivity, radiation dosages, drilling industry, hazard index.*

INTRODUCTION

Naturally occurring radioactive materials (NORMs) are mostly nuclides with the half-life of hundreds millions of years. Natural radioactivity is the major source of radiation to which humans are exposed and is responsible for more than 75% of all ionizing radiation (1). Also, exposure to background natural radiation (2.4 mSv/person/year) accounts for approximately 80% of the total radiation dose (2). Radioactive isotopes occur naturally in the environment but can accumulate due to industrial activities, so NORMs can be found in several industries, such as mining and milling activities, ore processing, cement production and petroleum industry (3, 4). Recently, more attention is given to occupational health hazards in petroleum industries, due to higher exposure rates. In such industries NORMs waste includes ^{238}U, ^{235}U, ^{232}Th and etc. These materials are brought to the earth surface in the fossil fuel exploration and extraction processes and radioactivity levels may exist above the background radioactive levels (5). The dominating radium radionuclides, ^{226}Ra and ^{228}Ra, range from 1 to 1000 kBq/kg (6).

Several studies have measured the natural radioactivity of oil and gas exploration wastes in the world (5, 7-11). The Khuzestan province, in southwest in the Iran, is rich in oil and gas areas. The aim of this study was to measure the levels of radioactivity in samples of petroleum drilling processes and also the activity concentrations of the ^{232}Th, ^{226}Ra and ^{40}K, in the samples in the area. The absorbed dose rates, radiation hazard indices and radium equivalent activity of gathered samples were calculated for sampling locations. Thus, evaluation of the extent of radiation exposure on these sites is of the utmost importance.

MATERIALS AND METHODS

In total, ten soil samples were randomly taken from different drilling areas in Khuzestan province. Figure 1 shows the map of sampling site of drilling areas. 2 Kg soil for each sample was picked up from the drilling cutting waste. The regions of samples are shown in table 1. In order to measure the radioactivity, soil samples were prepared according to the standard method ASTM C999 [11]. As mentioned in the method, soil samples were placed in an oven (Jeiotech model OF-01E, South Korea) with a temperature of 110 ° C for 12 hours to dry completely. In the next step, the specimens were placed in ball mill (Retsch MM-400, Germany) with ceramic balls for 1 night and then to obtain uniform and homogeneous powder, each sample was sieved with a 500µm mesh (US.NO.35). Next, the samples were placed in 500 mL marinelli beakers, closed tightly and stored in a cool place for at least four weeks. The activity concentrations were measured as follows, the samples were analysed with gamma-ray spectrometry with high purity germanium (HPGe) detector (GC 2020-7500) (CANBERRA XtRa, USA). The detector has a relative efficiency of 20%, resolution of 2 keV for 1332 kev photons of ^{60}Co. A multichannel analyser card (MCA) was installed in a PC computer for analysis purposes. The RGU standard sources (for U calibration and its chain elements), RGTh (for Th calcification and its chain elements) and RGK (for K calibration) were used for purpose of calibrations. Minimizing the background radiation is vital in gamma spectrometry, for this purpose, large lead shields with polyethylene layers were used. The time duration for each sample counting was 86400 s. a distilled water sample spectrum in the same geometry was used as background correction which was subsequently subtracted from each spectrum.

The equation (1) was used to determine the activity concentration (A) of each radionuclide [1].

$$A(Bq/g) = \frac{C}{\varepsilon y m} \qquad (1)$$

Where, C: the full-energy peak count rate for the radionuclide of interest (in counts per second), Ɛ: the amount of efficiency of detection for the specific energy, y: the correspondent gamma-ray yield, and m: the sample mass (gr).

The radiation emitted from environmental radionuclides is called the absorbed dose rate. This factor, $D(nGy/h)$ in air at the height of 1m above the ground level with regard to the concentrations of ^{226}Ra, ^{232}Th and ^{40}K was calculated according equation (2) [12]:

$$D_{\gamma r}=(0.427C_{Ra}+0.662C_{Th}+0.043C_{K})\times 10^{-3} \qquad (2)$$

C_{Ra}, C_T and C_K are the concentrations in Becquerel per kilogram of ^{226}Ra, ^{232}Th and ^{40}K, respectively [15,16]. As the recommended value is 55 nGy/h by UNSCEAR, $D_{\gamma r}$ must be lower than it [1].

The annual effective dose equivalent (AEDE) can be obtained from the equation (3) [1]:

$$AEDE(mSv/y)=(D(nG/h)\times 0.7(Sv/G)\times 0.2\times 8760)\times 10^{-6} \qquad (3)$$

In order to examine the health outcomes of the absorbed dose rates, The AEDE should be calculated. As the UNSCEAR [1] reports, a value of 0.7 Sv/Gy was used as the conversion coefficient arising from absorbed dose in the air to effective dose received by humans and another 0.2 value for the outdoor occupancy factor.

Total activities of materials that included ^{232}Th, ^{226}Ra and ^{40}K, was calculated by the radium equivalent index $^{226}Ra_{eq}$ was calculated according equation (4) [1].

$$^{226}Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{K} \qquad (4)$$

Where C_K, C_{Th} and C_{Ra} represent the activities of ^{40}K, ^{232}Th and ^{226}Ra (^{238}U-series) (Bq/kg) respectively.

The external hazard index (H_{ex}) is defined to limit the external γ -radiation dose and calculated by equation (5) [13].

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$$H_{ex}=C_{Ra}/370+C_{Th}/259+C_K/4810\leq 1 \quad (5)$$

The internal hazard index (H_{in}) which is obtained from the equation (6), measures the internal exposure to radon and its daughter products [13].

$$H_{in}=C_{Ra}/185+C_{Th}/259+C_K/4810\leq 1 \quad (6)$$

The level of γ-radiation hazard associated with the natural radionuclides can be estimated by another activity utilization index which is evaluated using this equation (7) [14].

$$I_\gamma=A_{Ra}/150+A_{Th}/100+A_K/1500\leq 1 \quad (7)$$

Where A_K, A_{Ra} and A_{Th} are the activity concentrations of and ^{40}K, ^{226}Ra and ^{232}Th respectively, in Bq/ kg for the samples. Iγ should be less or equal to 1.

Statistical analysis

Descriptive statistics and *t-test* were used to present the data. SPSS version 19 and Excel version 2013 were employed for data analysis. P value was considered less than 0.05.

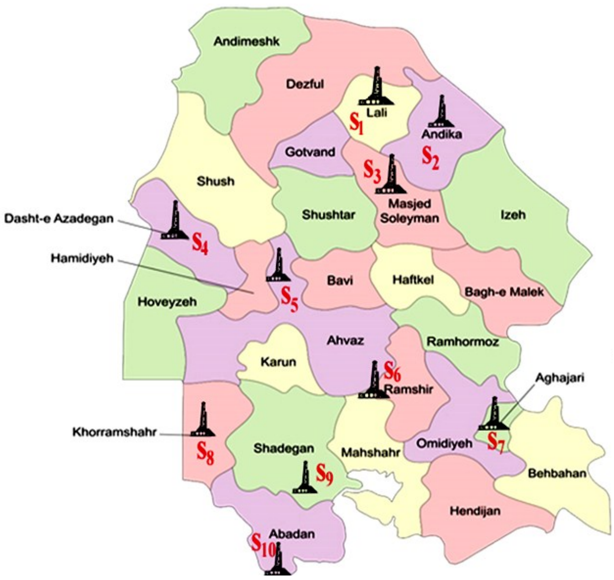

Figure1. The map showing the study area and sampling sites of drilling areas.

Table 1. The activity concentration of ^{232}Th, ^{226}Ra and ^{40}K of samples

Sample	activity concentration (Bq/Kg)		
	^{232}TH	^{226}Ra	^{40}k
s_1	19.223	45.006	305.21
s_2	12.527	34.905	427.333*
s_3	13.92	49.633*	310.797
s_4	22.054	40.655	392.202
s_5	23.8	49.993*	416.955*
s_6	17.015	33.255	334.656
s_7	16.19	44.998	346.347
s_8	20.299	30.897	96.88
s_9	4.994	17.907	52.55
s_{10}	11.234	19.596	81.75
Mean	16.126	36.684	276.468
World average	35	45	412

*one sample t-test results showed significant difference, P<0.05

RESULTS

Table 1 shows the radioactivity concentrations of naturally originated radionuclides of ^{232}Th, ^{40}K and ^{226}Ra in samples. According to UNSCEAR values for ^{232}Th (50 Bqkg^{-1}), ^{40}K (50 Bqkg^{-1}) and ^{226}Ra (500 Bqkg^{-1}), concentration in all soil samples were lower than the recommended values [1]. The maximum concentrations of ^{232}Th and ^{226}Ra in soil samples are reported for sample No.5, with activity concentration of 23.8 and 49.99 Bq/Kg respectively. Also, maximum concentration of ^{40}K (427 Bq/Kg) belongs to sample number 2. The minimum concentration of ^{232}Th, ^{226}Ra, and ^{40}K are 4.99 Bq/Kg, 17.90 Bq/Kg for (s_9) and 52.59 Bq/Kg for sample No.9, respectively. Table 1 also shows that only 40% of soil samples have activity concentrations of ^{226}Ra higher than the world average value [1]. The case is just 20% ^{40}K in soil samples be higher than average value.

Table 2 reveals the results of absorbed dose rate, AEDE and $^{226}Ra_{eq}$. Calculated gamma absorbed dose rate showed that all values are lower than the recommended value 55 nGy/h. The minimum value of the total absorbed dose rate was 13.21 nGy/h in sample 9. And the maximum value was 55.03 nGy/h in sample 5 (table 2). The annual effective dose equivalent varied from 0.016 to 0.067 mSv/y (table 2). The maximum and minimum of $^{226}R_{aeq}$ belonged to sample 5 (116.13 Bq/Kg) and sample 2 (29.04 Bq/Kg), respectively. The hazard indices (H_{ex} and H_{in}) for samples were calculated and are shown in table 2. According to the obtained values, the hazard indices were less than one unit. Also, results of the obtained values of Iγ for samples were less than one unit. A comparison of ^{232}Th, ^{226}Ra and ^{40}K concentrations of samples from various regions of the world are given in table 3. Figure 2 shows the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for all samples. Here the values are shown related to critical limit value.

Table 2. Results of the absorbed dose rate (D), the annual effective dose equivalent rates (AEDE), the radium equivalent activity (Ra_{eq}), the index of external and internal radiation hazard (H_{ex}, H_{in}) and Activity utilization index(I_γ).

sample	D(nGy/h)	AEDE(mSv/y)	Ra_{eq}(Bq/Kg)	H_{ex}≤1	H_{in}≤1	I_γ≤1
s_1	45.067	0.055	95.996	0.259	0.381	0.696
s_2	41.573	0.051	85.723	0.231	0.326	0.643
s_3	43.773	0.054	93.470	0.252	0.387	0.677
s_4	48.824	0.060	102.392	0.276	0.386	0.753
s_5	55.032	0.067	116.132	0.314	0.449	0.849
s_6	39.854	0.049	83.355	0.225	0.315	0.615
s_7	44.825	0.055	94.818	0.256	0.378	0.693
s_8	30.797	0.038	67.384	0.182	0.265	0.473
s_9	13.212	0.016	29.095	0.079	0.127	0.204
s_{10}	19.320	0.024	41.955	0.113	0.166	0.297
Mean	38.23	0.047	81.03	0.22	0.32	0.59

Table 3. Comparison of values for ^{232}Th, ^{226}Ra and ^{40}K concentrations of samples from various regions of the world

Country	Activity Concentration (Bq kg^{-1})			Radiological parameters						References
	^{232}Th	^{226}Ra	^{40}K	D (nGy/h)	AEDE (mSv/y)	Ra_{eq} (Bq/Kg)	H_{ex}≤1	H_{in}≤1	I_γ≤1	
Turkey	83.1	79.3	1273.7	208.5	1.02	232.8	0.63	0.84	0.86	(10)
Greece	107.6	74	88.1	-	-	-	0.14	0.19	0.96	(9)
Malaysia	52	-	610.8	74.8	0.92	-	0.44	0.19	0.96	(20)
Brazil	107.6	72.8	1127.1	-	-	313.5	0.55	-	1.16	(8)
Turkey	64.7	78.9	238.4	86.1	0.42	189.9	0.51	0.73	0.67	(21)
Iran (Golestan)	31	23	453	50	61.4	102.4	0.28	0.34	-	(19)
World average	45	32	412	-	-	-	-	-	-	(22)
Iran	16.2	36.2	276.4	38.23	0.047	81.03	0.22	0.32	0.59	Present study

DISCUSSION

This study is one of the first attempts in order to assess radionuclides activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in soil samples taken from oil drilling cutting waste in Iran, Khuzestan province. The mean activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th were 276.468, 36.684 and 16.126 Bq.kg^{-1}, respectively. Which ^{226}Ra was higher than the amounts reported for Iran (Golestan (23 Bq. kg^{-1})) and the average amount reported for the world in general but it was lower than the other parts of the world [8-10, 19-22]. In a study by Mouandza *et al.* (2018) the results showed that 74% of measured area had activity concentrations of ^{226}Ra higher than world average value [23], however the present study reports this difference for about 40% of samples. This different value can be explained by this fact

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that in Mouandza study the samples were taken from a location uranium main. Also, for the case of ^{232}Th, our findings showed a lower amount than any other studies (8-10, 19-23). On the other hand the measured values for ^{40}K, were lower than all mention studies except Turkey (238.4 Bq.kg^{-1}) and Greece (88.1 Bq.kg^{-1}) (9, 21). The mean activity concentrations are lower than the world mean values identified by UNSCEAR for ^{40}K and ^{232}Th (1). The average absorbed dose rates (D), Radium equivalent activity (Ra_{eq}) and annual effective dose equivalent (AEDE) and various hazard index (H_{ex}, H_{in} and $I\gamma$) for samples were calculated and were 38.228 nGy/h, 0.047 mSv/y, 81.032 Bq/Kg, 0.219, 0.318 and 0.59, respectively. All of the above mentioned values are below the permissible limit.

CONCLUSION

Present study has analyzed the natural radioactivity content of ten different soil samples of oil drilling cutting wastes for the measurement of radioactivity. The findings of this study demonstrated that all the calculated values are below the recommended maximum values in the UNSCEAR reports, but were higher than the world average values in some cases. It can be concluded that it is safe for workers who are working in oil and gas drilling sites in these regions of Khuzestan province, in terms of radiation hazards. This study could be used as a track for further investigations and this data might be useful for the naturally occurring radioactivity mapping.

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