Estimation of the Radiation Dose for Some Individuals Working With Naturally Occurring Radioactive Materials

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Abstract: The aim of the present study is to estimate the radiation dose for some individuals who are working in oil companies. Radiation hazard arises due to high content of naturally occurring radioactive materials (NORM) in the field of work. The radiation workers under investigation were examined externally by using thermoluminescence detectors such as TLD-100 (LiF7 (Mg, Ti)) for assessing the external dose and internally by using whole body counter with NaI(Tl) detector to evaluate the internal dose. The present results indicated that there is no any internal contamination. Total body potassium TBK content was detected and ranged from 1.59 to 2.59 g K/kg. In addition; the annual effective dose resulting from ^{40}K varied from 174 to 287 μSv. On the other hand, the external dose measured using TLD-100 dosimeters ranged from 0.38 to 0.73 mSv/y. These values are below the published worldwide limits according to latest ICRP publications (ICRP 103, 2007). Finally, the activity concentration of the analyzed NORM samples by using HPGe detector is 187 kBq/kg for Th-232, 607 kBq/kg for U-238 and 416 kBq/kg for Ra-226.

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Key word: NORM; TLD; External dose; Internal dose; Whole body counter.

1. Introduction:

According to the exposure pathway of natural radiation, the public dose for humans can be divided into external and internal exposures. External exposure comes from the exposure to cosmic rays and terrestrial gamma rays. Internal exposure comes mainly from radon progeny and potassium-40 (DOE 1998). The human population is always exposed ionizing radiation due to background radiation. Besides man-made radiation, the main source of background radiation is natural radioactivity. Natural radioactivity has existed since the beginning of the universe due to long half-lives of the natural radioelement found in the earth's crust. These radionuclides of Ra-226, Th-232 and K-40 can be found almost in all types of rocks, granite, sand, cement and gypsum from which building materials are produced (Yu-Ming Lin et al., 1996). As a result of oil and gas production and processing operations, naturally occurring radioactive materials (NORM) accumulate at elevated concentrations in by-product waste streams. The sources of most of the radioactivity are isotopes of uranium-238 and thorium-232, which are naturally present in the subsurface formations from which oil and gas are produced. The primary radionuclide of concern in NORM wastes is ^{226}Ra, of the ^{238}U decay series. Radium-228, of the Th-232 decay series, also occurs in NORM waste but is usually present in lower concentrations (Mavi B and Akkurt I, 2010).

The production waste streams most likely to be contaminated by elevated radium concentrations include produced water, scale, and sludge. Radium, which is slightly soluble, can be mobilized in the liquid phases of a subsurface formation and transported to the surface in the produced water stream. Dissolved radium either remains in solution in the produced water or, if the conditions are right, precipitates out in scales or sludge.

Scales and sludge removed from production equipment often are disposed of by land spreading, a method in which wastes are spread over the soil surface to allow the hydrocarbon component of the wastes to degrade.

Under existing regulations for workers classified as radiation workers, doses are required to be as low as reasonably achievable, not to exceed an annual dose of 20 mSv. This limit would apply to workers who handle NORM only if they were classified as radiation workers by ICRP regulations; otherwise (ICRP 103, 2007), NORM workers are subject to dose limits that apply to the general public. The currently accepted public dose limit recommended by the International Commission on Radiological Protection is 1 mSv/y from all sources.

The objective of this study was to estimate potential radiological dose to 10 workers during maintenance and radiation decontamination of the petroleum equipments resulting from the disposal of NORM wastes.

2. Materials and methods

2.1. Measuring equipments

The current work was carried out using three different techniques. These techniques are installed at the Nuclear Research Center (EAEA) and they are as following:

1. Low level radioactivity measurement using high purity germanium detector for measuring radioactivity concentration of environmental samples,
2. Whole body counter FASTSCAN based on two vertical NaI(Tl) detector to measure the internally radiation contaminants, and
3. Harshaw 6600 reader for reading the registered external dose.

3. Sample and individual preparation for measurement

3.1. NORM Sample preparation

Samples of NORM were taken from waste associated with oil and natural gas production. The waste bulk contained the sludge and scale formed inside the production equipment (pipelines, tank separators, pumps). These wastes were removed periodically during the maintenance process. For measuring activity concentration, 250 g (0.25 kg) of waste sample was packed in a plastic container (typical use for calibration purposes), sealed and stored for 4 weeks to establish the secular equilibrium between the parent radionuclides and daughters (^{226}Ra and ^{222}Rn).

3.2. Individual preparation for measuring

For in vivo measurements individuals should be free from external contamination and in fresh clothing, take shower, wash hair, often disposable paper garments before entering the monitoring area. Accessories such as jewellery, watches and spectacles should be removed. Such precautions help to avoid false identification of internal activity, and also prevent the transfer of contamination to the counting equipment. The subject stands inside a shield and on an axis parallel with the detectors.

4. Used equipment

4.1. Gamma ray spectrometry

The gamma ray measurements for the sludge samples were carried out using High Purity Germanium (HPGe) detectors (model Canberra) with relative efficiency of 25%, the correlation between energy to channel number was 0.5 keV/ch. The gamma ray spectra were analyzed by using a software program (Genie 2000) which calculates the activity concentration of the samples. The natural background level was subtracted from the accumulated spectrum.

4.2. Whole Body Counter (WBC)

WBC uses two large NaI (Tl) detectors. Each detector has dimensions of 3"x 5"x 16", configured in a linear array on a common vertical axis. The detectors are connected to a single photo-multiplier tube. WBC is gamma-ray spectrometers which used for the detection and quantitative measurement of radioactivity within the human body. WBC system uses Canberra's ABACOS and Genie 2000 Software package to analyze the obtained gamma spectra and calculate the radioactivity concentration in different duration times. WBC system is a powerful tool for measuring the internal contamination for occupational in nuclear power plants, nuclear facilities and activities related directly to the emissions of gamma rays and might be inhaled or ingested.

4.3. TLD material

The TLD-100 contains natural lithium with about 7.5 percent of the lithium ions being ^{6}Li and the remainder ^{7}Li. The basic dosimeter, a Harshaw TLD-100 holds two LiF7 crystals, the second one being filtered in order to correct for energy response. The information of dose exposure is provided by a card reader, model Harshaw 6600. It has the capability to heat two TLDs at the same time, thus providing a luminescence response proportional to irradiation. The luminescence light is detected by a photomultiplier tube, whose current signal, integrated on time, gives the dosimeter reading (nC). The signal is assumed to be linear with dose.

5. Used equations

5.1. Calculation of absorbed dose rate from NORM samples

The absorbed dose rate in nGy/y due to gamma ray at 1m from NORM sample was calculated, according to the equation (UNSCEAR 2000):

$$D = R_{Ra}C_{Ra} + R_{Th}C_{Th} + R_K C_K \qquad (1)$$

where; R_{Ra}, R_{Th} and R_k, are the conversion factors in (nGy/h)/(Bq/kg) of ^{226}Ra **,** ^{232}Th and ^{40}K; C_{Ra}, C_{Th} and C_{40} are the activity concentrations in Bq/kg of ^{226}Ra **,** ^{232}Th and ^{40}K. In the UNSCEAR recent reports (2000), the Committee used 0.7 Sv/Gy for the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.2 for the outdoor occupancy factor. Effective dose rate in air outdoors in units of µSv/y is calculated by the following formula:

Effective dose rate (µSv/y) = Dose rate (nGy/h) × 24 h × 365.25 d ×0.2 (occupancy factor) ×0.7 Sv/Gy (conversion coefficient) × 10^{-3} (2)

The dose conversion factors are used to convert the activity concentrations of U-238 (Pb-212) series and Th-232 (Pb-214) series into doses (nGy/h per Bq/kg)) as 0.04342 and 0.01917 (UNSCEAR, 2000) and for Ra-226 is 0.00108 (Tzortzis M et al 2003).

As long as the workers in this field carry out this operation 4 times per year and the duration time for one operation is 6 hours so the total duration time for all operations is 24 hours (one day). On the light of this data equation number 2 can be rewritten as following:

Effective dose rate (μSv/y) = Dose rate (nGy/h) × 24 h × 1 d ×0.2 (occupancy factor)×0.7 Sv/Gy (conversion coefficient) × 10^{-3} (3)

And the effective dose rate per operation is calculated as following:

Effective dose rate per operation (μSv) = Dose rate (nGy/h) × 6 h ×0.2 (occupancy factor)×0.7 Sv/Gy (conversion coefficient) × 10^{-3} (4)

The working hours per year 2000 (83 day per year) and if the worker is carrying out the operation every working days so the effective dose rate per year can be calculated from the following equation:

Effective dose rate (μSv/y) = Dose rate (nGy/h) × 2000 h × 0.2 (occupancy factor)×0.7 Sv/Gy (conversion coefficient) × 10^{-3} (5)

5.2. Estimation of the Annual Effective Dose from ^{40}K

From (ICRP 2, 1978); and for ^{40}K it can be found that the effective absorbed energy per disintegration is 0.463 MeV for the β-particles and 0.091 MeV for the γ – rays. From the natural long half-life isotope of the K present in the human body; the adult man (70 kg) contains about 140 g of potassium and the 0.0118 % abundant isotope ^{40}K corresponds to 4400 Bq with 11% probability of gamma ray emissions.

The annual dose is given by equation:

$$D = \frac{q\,.\,t\,.\,\varepsilon\,.\,\alpha}{m} \qquad (6)$$

where q (Activity) = 4400 Bq, t (exposure time) = 3.15 ×10^{7} s/y, ε = 0.554 MeV,
α = 1.6 ×10^{-13} J/MeV and m = 70 kg (is the body mass for reference man).

Then from the last equation, the annual effective dose can be calculated for the reference man resulting from ^{40}K and found to be 176 μSv.

6. Results and discussion

6.1. Radioactivity measurement and dose estimation for NORM sample

As shown in figure 1 the γ-ray spectrum of one NORM sample; it is obvious that the main radionuclides in the NORM sample are U-238 and Th-232.

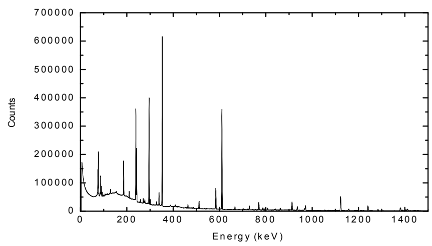

Fig (1) gamma ray spectrum for NORM sample

Table 1 shows the activity concentration is shown in the calculate values of absorbed dose and annual effective dose are shown too in Table (1).

Using absorbed dose conversion factors ADCF (nGy/h per Bq/kg) calculated by Clouvas A. et al., 2000 and the specific activity concentration (Bq/kg) of Ra-226, U-238 and Th-232 measured using HPGe gamma spectrometer for the studied samples the effective dose µSv was calculated per operation, per 4 operations and per year as shown in figure 2.

Table (1) the results of the analyzed sample of NORM using HPGe detector

Radionuclide	Activity kBq/kg	Absorbed dose rate µGy/h	Effective dose (µSv)		
			per operation	per 4 operations	per year
(^{232}Th series)	187	3.58	3	12	1004
(^{238}U series)	607	26.36	22	89	7380
^{226}Ra (186 keV)	416	0.45	0.4	1.5	126
Total	**1210**	**30.39**	**25.4**	**102**	**8510**

For ^{232}Th activity concentration was determined by measuring the energy lines for gamma ray from ^{212}Pb. ^{238}U activity concentration was determined by measuring gamma ray of ^{214}Pb energy lines and the activity concentration for ^{226}Ra was determined by measuring the gamma ray resulting from the energy line 186 keV. It is obvious from Table (1) that the activity concentration from samples is higher than the recommended limit (45 Bq/kg, 33 Bq/kg and 32 Bq/kg for Th-232, U-238 and Ra-226 respectively) (UNSCEAR 2000). Also, the absorbed dose rate from the studied petroleum samples is higher than the recommended value; 59 nGy/h (UNSCEAR 2000). Finally, the annual effective dose of the studied sample to a worker from occupational exposure 8.5 mSv is lower than the recommended value for occupational (20 mSv).

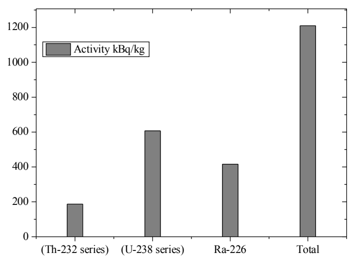

Fig 2.a. Activity concentration in kBq/kg

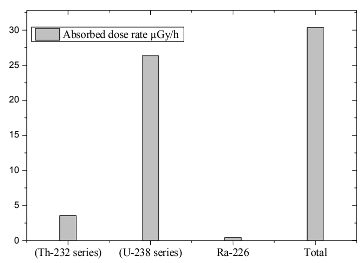

Fig 2.b. Absorbed dose rate in µGy/h

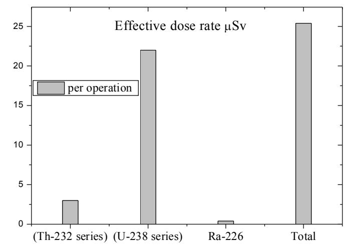

Fig 2.c. Effective dose rate µSv/one operation

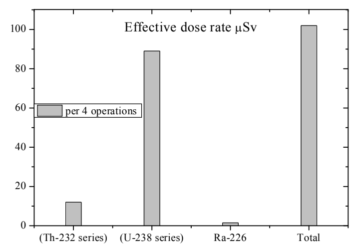

Fig. 2.d. Effective dose rate in µSv/4 operations

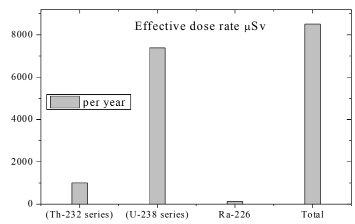

Fig. 2.e. Effective dose rate in µSv/y.
Figure (2) Activity, absorbed dose and effective dose rate per different times

6.2. Internal dose using WBC

The obtained results from whole body counter (WBC) measurements for 10 individuals indicated that there is no any internal contamination but found that the activity resulting from body burden ^{40}K ranged from 4.376 to 6.309 kBq/ body mass with an average activity of 5.564 kBq/ body mass, in addition to the annual internal dose from ^{40}K ranged from 174 to 287 µSv with an average dose of 246 µSv. The current results for internal dose from ^{40}K are in good agreement with previous results measured by Morsi TM, 2011. Table (2) presents the internal dose and TBK content for 10 individuals working with naturally occurring radioactive materials (NORM).

6.3. Thermoluminescent dosimetry measurement

In this work, the present results obtained by using TLD show that the external dose rate ranged from 0.38 to 0.73 mSv/y. These values are acceptable compared to the ICRP recommendation (1 mSv/y for public and 20 mSv/y for worker) (ICRP 103, 2007). Table (3) represents the external dose rate for workers dealing with naturally occurring radioactive materials.

Table (2) internal dose and TBK content for some individuals

Subject	Activity of ^{40}K (Bq)	TB K (g)	Internal dose µSv/y	
			^{40}K	Other radionuclides
Case 1	6163	202	276	ND*
Case 2	6309	207	282	ND
Case 3	6260	205	252	ND
Case 4	4885	160	287	ND
Case 5	5409	177	225	ND
Case 6	5224	171	256	ND
Case 7	5358	176	218	ND
Case 8	6251	205	280	ND
Case 9	4376	143	174	ND
Case 10	5403	177	215	ND

*ND= Not Detectable

Table (3) external dose for 10 individuals using TLD-100

Subject	External dose (µSv/y)
1	440
2	480
3	500
4	420
5	730
6	480
7	380
8	460
9	370
10	720

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7. Conclusion

The average annual effective dose resulting from the studied sludge sample was found to be 8.5 mSv, this value is lower than 20 mSv compared to the dose limit for radiation workers. The obtained results fulfill to those recommended by ICRP publication 103 (2007). In case of the internal radiation dose, the results of scanning using WBC for counted individuals indicated that there is no internal contamination. Activity concentration for the NORM samples was higher than the worldwide published value.

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