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Investigation of Naturally Occurring Radioactive Materials in Sudan Petroleum Industry

R. S. Abdalraheem[1] *, I. Salih*[1] *and S. Takriti*[2*]

[1]Sudan Atomic Energy Commission (SAEC).
[2]Arab Atomic Energy Agency (AAEA), Tunis, Tunisia.

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Abstract: The oil and gas industry are one of the industries that exposed workers are at risk of radioactive materials concentrated by the industry, as it accompanies radioactive materials natural oil extracted from the ground, to be placed on the interior walls of pipes transporting oil and stores the chapter or stores stored, these depositions are Scale or Sludge.
Surveys were conducted for evaluations of radioactivity levels in this area at different parts of Sudan where the production and exploration of oil is conducted. A set of 29 samples was collected from two areas: 12 samples from the refinery AL-jaily and 17 samples from the area of Heglig and the neighboring areas. The levels of natural radioactivity varied with uranium concentration taking values between 188 - 250 (Bq/Kg) in Al-jelly refinery and 80-25200 (Bq/Kg) in the Heglig area. Neighboring regions' results showed relatively higher uranium concentrations in the areas of oil exploration and drilling and also in oil filter samples.
Keywords: NORM, Radioactivity, Oil sludge waste.

1 Introduction

Naturally occurring radioactive material (NORM) can concentrate as the result of human activities such as oil and gas operations, mining, water treatment, etc. However, there are situations where exposures to natural sources may warrant consideration as to whether controls should be applied (1). One such situation is where the conditions are conducive to the buildup of elevated concentrations of radon in the air. Another situation is the mining and/or processing of material where the activity concentrations of radionuclides of natural origin in the material itself, or any material arising from the process, are significantly elevated - such material has come to be referred to as Naturally Occurring Radioactive Material (NORM). (2)

Radioactive materials such as Uranium and Thorium were incorporated in the Earth's crust when it was formed; these normally exist at trace (parts per million – ppm) concentrations in rock formations. Decay of these unstable radioactive elements produces other radionuclides that, under certain conditions (dependent upon pressure, temperature, acidity, etc) in the subsurface environment are mobile and can be transported from the reservoir to the surface with the oil & gas products being (3).

In the past, regulatory attention has been focused mostly on exposures arising from the mining and processing of uranium ores because such activities are part of the nuclear fuel cycle. More recently, attention has been broadened to include exposures from other industrial activities involving NORM, in recognition of the potential for such activities to also give rise to significant exposures of workers and members of the public if not adequately controlled.

More and more countries are now including provisions in their national legislation and regulations for the control of exposures to natural sources, and the body of radiological data on such exposures is growing rapidly. This material, when technologically enhanced (TE), can present serious health and safety hazards if it is not handled and disposed of properly. TENORM associated with uranium, thorium, and their associated decay series is of the greatest concern.
TENORM responses are evaluated to decide if a radioactive materials license is required for possession of the material. There is currently no satisfactory cost-effective way of disposing of this material. Waste brokers, such as Envirocare, will take the material at a substantial cost.

There are three types of radiation emitted by NORM, namely;
• Alpha (α), Beta (β) and Gamma (γ)

*Corresponding author e-mail: stakriti@gmail.com

Alpha particles are helium nuclei that are heavy and doubly (positively) charged which causes them to lose their energy very quickly in matter. They can be stopped by a sheet of paper or the surface layer of your skin. Alpha particles are considered hazardous to a person's health only if a radioactive source of alpha-emitting particles is inhaled or ingested.

Beta particles are much smaller and only have one (negative) charge, which causes them to interact more slowly with the material. They are effectively stopped by thin layers of metal or plastic and are again considered hazardous only if a beta emitter source is ingested or inhaled.

Gamma emitters are associated with alpha, and beta decay and are a form of high-energy electromagnetic radiation that interacts lightly with matter. Gamma rays are best shielded by thick layers of lead or other dense materials and are considered an external hazard to living tissues (i.e. the human body) (3).

This study aims to determine how uranium is transferred from phosphate in the sedimentary basins of Tunisia to solid-liquid waste in addition to illustrating the radiation ratio of uranium in solid and water waste to determine the equilibrium time of phosphate layers containing uranium.

2 Materials and Methods

1- Gamma-ray Spectroscopy

Gamma-ray spectrometry is an analytical method that allows the identification and quantification of gamma-emitting isotopes in a variety of matrices. In one single measurement and with little sample preparation, gamma-ray spectrometry allows you to detect several gammas emitting radio nuclei in the sample. Radioactive nuclei commonly emit gamma rays in the energy range from a few keV to ~10 MeV, corresponding to the typical energy levels in nuclei with reasonably long lifetimes. Such sources typically produce gamma-ray "line spectra" (i.e., many photons emitted at discrete energies), whereas much higher energies (upwards of 1 TeV) may occur in the continuum spectra observed in astrophysics and elementary particle physics. The boundary between gamma rays and X-rays is somewhat blurred, as X-rays typically refer to the high energy EM emission of atoms, which may extend to over 100 keV, whereas the lowest energy emissions of nuclei are typically termed gamma rays, even though their energies may be below 20 keV.

Most radioactive sources produce gamma rays of various energies and intensities. When these emissions are collected and analyzed with a gamma-ray spectroscopy system, a gamma-ray energy spectrum can be produced. A detailed analysis of this spectrum is typically used to determine the identity and quantity of gamma emitters present in the source. The gamma spectrum is characteristic of the gamma-emitting nuclides contained in the source, just as in optical spectroscopy, the optical spectrum is characteristic of the atoms and molecules contained in the sample.
The equipment used in gamma spectroscopy includes an energy-sensitive radiation detector, a pulse sorter (i.e., multichannel analyzer), and associated amplifiers and data readout devices. The most common detectors are detectors include sodium iodide (NaI) scintillation counters and high-purity germanium detectors.

2- Sampling and Measurements

Sampling and sample preparations were conducted according to guidelines specified by IAEA in their reference book (4). In brief, this is as follows:
Samples received in the laboratory may not be in the proper physical form for analysis. They may require size reduction, drying, and some form of homogenizing before aliquots can be taken for analysis.

After collection, the samples were properly stored to avoid degradation, spoiling, or other decomposition. Proper care must be taken to avoid loss of volatile radionuclides. Short periods of storage before analysis may require refrigeration, freezing, or the addition of a preservative. When long periods of storage are needed, it may be preferable to convert the samples to a more stable form immediately after sampling. The sample collection equipment, containers, and sample preparation areas must be kept clean to avoid contamination.

Environmental samples of low-level radioactivity are often measured in Marinelli beakers. They are made with chemically resistant polypropylene and are available for gamma spectral analysis of a variety of liquid solutions or solid samples. These Marinelli beakers are advantageous for the following reasons:

1. Higher counting efficiencies.
2. Lighter weight which requires mini storage space
3. Seamless, thin-wall construction eliminates leakage and minimizes gamma ray attenuation.
4. Cost effective.

3- Detector Efficiency

Not all gamma rays that pass through a detector will produce a count in the system. The probability that a gamma ray will interact with the detector and produce a count is the efficiency of the detector. High-efficiency detectors produce spectra in less time than low-efficiency detectors. In general, larger detectors have higher efficiency than smaller detectors, although the shielding properties of the detector material are also an important factor. Detector efficiency is measured by taking a spectrum from a source of known activity and comparing

the count rates in each peak to the count rates expected from the known intensities of each gamma ray.

Efficiency, like resolution, can also be expressed in absolute or relative terms. The same units are used, percentages, so the spectroscopist must take care to determine which kind of efficiency is being given for the detector. Absolute efficiency values give the probability that a gamma ray of specified energy passing through the detector will interact with the crystal and be detected. Relative efficiency values are often used for Germanium detectors, and compare the efficiency of the detector at 1332 KeV to that of a 3"x3" NaI detector. Relative efficiency values greater than 100% can therefore be encountered when working with very large Germanium detectors.

The energy of the gamma rays being detected is an important factor in the efficiency of the detector. By plotting the efficiency at various energies, an efficiency curve can be obtained. This curve can then be used to determine the efficiency of the detector at energies different from those used to obtain the curve. Figure (1) shows plots of HPGe detectors respectively used in the current study. The calibration sources were obtained in two geometries; Marinelli beakers half and 500 kilogram. It contains the following radionuclides: Uranium, Thorium, and Potassium at different activities (5).

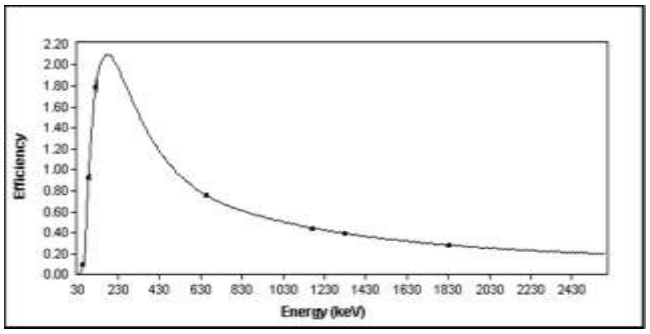

Fig.1: Efficiency (%) calibration curve for HPGe detector.

4- Reference materials

RGU-1, RGTH-1, and RGK-1 are intended for calibrating laboratory gamma-ray spectrometers for the determination of U, Th, and K in geological Materials. RGU-1 was prepared by the Canada Center for Mineral and Energy Technology (CANMET) under contract with the International Atomic Energy Agency. The material was prepared by dilution of Canada Certified Reference Material Project (CCRMP) Uranium ore BL-5(7.09%) with a floated silica power of similar grain size distribution BL-5 has been certified for Uranium, ^{226}R, and ^{210}P confirming that it is radioactive equilibrium. The complete description of the preparation and certification of RGU-1 may be found in the reference. (6).

Table 1: the energies, their respective branching ratios, and the corresponding efficiency of the radionuclides in the standard.

Radionuclide	Energy	CPS	I_η	Eff. E-5
U-238	63	0.1057	4.8	0.89
Th-232	92	0.1121	2.8	1.62
Ra-226	186	0.2634	3.59	2.97

Table (1) shows the energies, their respective branching ratios, and the corresponding efficiency of the radionuclides in the standard.

5- Radioactivity Measurement

The activity was calculated by the following formula:

$$A = \frac{N(cps)}{I_\gamma \cdot \eta \cdot m}(Bq \cdot Kg^{-1})$$

Where:

- A: absolute transition probability of gamma-decay (Bg/ Kg).
- $N(cps)$: Net counting rate of gamma-ray (counts per second).
- I_γ : Intensity of gamma energy (constant).
- m: the mass of the sample (Kg).
- η: Detector efficiency of the specific gamma–ray

3 Results and Discussion

The results of investigations in two areas (Al-jaily and Heglig) are tabulated in Tables (2) and (3). The tables show a concentration of naturally occurring radionuclides in each sample taken.
Samples were collected from two different regions of Sudan, where the production of oil and processing (refinery) are conducted. The study conducted at the AL-Jaily refinery showed the levels of uranium range between 188 and 250 Bq/Kg. At the oil exploration areas (Heglig, GNPO Concession) the levels of uranium range between 80 and 25200 Bq/Kg. The reason for the high concentration of radionuclides in the petroleum industry is attributed to the presence of water production with oil and the presence of scales and sleds.

Environmental radioactive contamination occurs in the oil fields in several ways including throwing water collection produced in the vicinity of the central stations and maintenance of reservoirs, and wells, and these practices will lead to contamination of soil and extend to large areas sometimes. The soil is contaminated with radioactive and according to international standards or local. Reaction, for example, determines the Syrian standards for soil contaminated with radium ^{226}Ra in the oil fields:

1 - Soil that increases the concentration of ^{226}Ra 0.15 Bq/g does not need to be addressed.
2 - Handle all soils that radiation increases the activity specific for Bq/g 5.2 as radioactive waste.
3 - Soil that is active radiation qualitative 0.15-5.2 Bq/g needs to address the on-site radiation exposure to fall to less than 0.1 mSv/y.
This pollution must be removed at the earliest possible opportunity to prevent the exposure of workers and the general public.

By comparing the concentrations in this work and radioactivity concentration in some countries the following can be observed:

The lowest activity concentration value was obtained from ^{324}Th (188 Bq/g) in the Oil field in Al-jaily and (2671 Bq/g) in Heglig, GNPO Concession. Compared with similar work in some countries the highest value obtained from the result obtained in the present study covers only a part of one region of the country.

The radionuclide ^{238}U, ^{232}Th and ^{40}K- in soil samples (Bq/Kg) from Kuwait, India, Egypt, Pakistan, Sir Lanka, Poland and Bangladesh are: (13.3 ± 5.0, 10.0 3.4 and332± 104),(15-71, 15-776 and 200-854),(165± 5 and71 ± 2 for ^{238}U and ^{232}Th), (9-40 and 7-105for ^{232}Th and ^{40}K) (72 and 585 for ^{232}Th and ^{40}K), (25-62 and 320-1200 for ^{232}Th and ^{40}K) and (36.7and458.2 for ^{232}Th and ^{40}K) respectively (7-13).

It is estimated that in areas of the normal radiation background, the world average values for ^{238}U, ^{232}Th, and ^{40}K activity concentrations in soil are 35.30 and 400 Bq/kg respectively. According to the UNSCEAR reports (1993, (14).

Table 2: The concentration level of radionuclide collected observed in Area (Al-Jaily oil refinery) (Bq/Kg).

Number	Sample code	Net count	Concentration
1	Kr7	0.0052	247
2	Kr8	0.0035	167
3	Kr14	0.0052	244
4	Kr24	0.0028	133
5	Kr31	0.0029	139
6	Kr2	0.0053	250
7	Kr18	0.0038	182
8	Kr25	0.0048	228
9	Kr26	0.0032	151
10	Kr29	0.0027	126
11	Kr35	0.0039	186
12	Kr1	0.0043	204

Max	25200
Min	80
Mean	2671
Sd	7029
Median	215

Table 3: The concentration level of the radionuclide collected observed in Area (Heglig, GNPO Concession) (Bq/Kg).

Number	Sample code	Net count	Concentration
1	N1	0.0045	213
2	N2	0.0295	138
3	N3	0.0037	178
4	N4	0.0626	293
5	N5	0.0053	251
6	N6	0.0055	258
7	N7	0.0034	162
8	N8	0.0045	215
9	N9	0.0025	120
10	N10	0.0172	808
11	N11	0.0075	352
12	N12	0.0028	131
13	N13	0.0051	243
14	N14	0.0017	080
15	N15	0.0538	25200
16	N16	0.0355	16600
17	N17	0.0036	172

Max	25200
Min	80
Mean	2671
Sd	7029
Median	215

4 Conclusions and Recommendation

These results are regarded as preliminary because adequate data describing the source term and other input parameters are not available. Absolute health risks cannot be determined from these preliminary estimates because of the uncertainty related to the source term concentrations and because of the conservative nature of many of the

input assumptions. For the same reasons, conclusions about the risk to human health should not be drawn by comparing the estimated doses with existing or proposed regulatory standards, such as dose limits and drinking water standards.

Given these limitations, it is still possible to draw some general conclusions regarding the selected NORM management and disposal methods and to compare the estimated doses as a preliminary indication of the relative risks associated with each method. The existing regulated standards can be used to provide a benchmark value that adds perspective to the results.

NORM wastes generated by the petroleum industry should be further characterized to improve efforts to assess potential doses to workers and the general public. Further characterization of the source term concentration is particularly important. Existing data collected at the state and company levels should be aggregated to improve efforts to calculate statistically representative source term concentrations.

Provided further assessment supports the results of this study indicating that subsurface disposal methods constitute realistically safe methods for disposing of NORM-contaminated wastes, state regulatory agencies should be encouraged to permit subsurface disposal projects more readily. Underground injection projects should not be considered to pose a significantly greater risk to the general public than downhole encapsulation projects. Regulators should strive to educate the public about the realistic risks related to subsurface disposal so that unfounded fears do not complicate the permitting process. Because this study did not consider the political, economic, sociological, and - radiological issues related to smelting NORM-contaminated equipment generated by the petroleum industry, further study is needed to fully examine the feasibility of this disposal option.

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