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Studying of Naturally Occurring Radioactive Materials (NORM) in Oilfield (A/100) South East of Libya

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Abstract: The huge volume of Naturally Occurring Radioactive Materials (NORM) wastes produced annually by the oil and gas industry in Libya deserves the attention of the national environmental protection authority, radioactive waste management and regulatory bodies. An investigation was carried out to find out the concentration of (NORMs) in evaporation ponds sludge in south eastern oilfield (A/100) of Libya. Twenty soil samples were collected from five evaporation ponds sludge. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil generated during oil production operations were determined using a gamma spectroscopy system based on High Purity Germanium (HPGe) detector. Concentrations ranged from 83 to 1000 Bq kg^{-1} for ^{226}Ra, 59 to 315 Bq kg^{-1} for ^{232}Th and 109 to 304 Bq kg^{-1} for ^{40}K. To evaluate the radiological effects, radium equivalent activity and external hazard are calculated. The magnitude of these results demonstrates the need of screening oil residues for their radionuclide content in order to decide about possibility of minimize the environmental impact of NORM and their final disposal. Disposal of NORM waste has to be in accordance with national regulations, environmental policy and international agreements and conventions. The researchers recommend limits for clearance and disposal, based on best international practice.

INTRODUCTION

Libya is a member of OPEC and one of the largest crude oil exporters of the world. Oil reserves in Libya are the largest in Africa. According to Oil and Gas Journal (OGJ). In January, 2013, Libya has proved crude oil reserves of 48 billion barrels. This accounts for about 38% of the total continent's reserves and the ninth largest in the world. Oil production was 1.7 million barrels per day (270×10^3 m^3 d^{-1}) as of 2010. Libya is considered a highly attractive oil area due to its low cost of oil production, proximity to European markets and of high quality because of its low sulphur content. Recently, serious attention has been given to health impacts from the uncontrolled release of waste containing TENORM, which stands for technologically enhanced naturally occurring radio nuclides which concentrated and accumulated in tubing and surface equipment in the form of scale and sludge (IAEA, 2003a, b; Hamlat *et al*., 2002). Produced water is water trapped in underground formation that is brought to surface along with oil or gas production and usually separated from oil and disposed, e.g., in an injection well or disposal well. Some companies may discharge this water into the environment for evaporation. Unlined ponds are usually built to collect this water and become highly contaminated with NORM (contaminated soil as this case of study). Management of produced water and its environmental effects, present challenges to the oil industry and environmental experts in Libya (Hamid *et al*., 2008; Massuod *et al*., 2012). Many studies worldwide reveal the impact of NORM in oil and gas production (Shawki *et al*., 2001; Al-Masri and Suman, 2003; Hamlat *et al*., 2003; Khodashenas *et al*., 2012). Equipment contaminated with NORM cannot be released for sale or disposal without being decontaminated and verified free of NORM contamination. Waste material contaminated with enhanced levels of NORM has to be disposed of in a controlled manner to ensure that it does not provide an unacceptable risk to the environment and the general public. In Libya, this problem has not been investigated in detail. This study presents results of analysis of contamination soil samples from ponds in south eastern oilfield of Libya and reviews different regulations implemented by countries dealing with soil contamination by NORM. The authors recommend limits for clearance and disposal, based on best international practice.

MATERIALS AND METHODS

Study area and sample collection: In this study all samples were collected from oilfield (A/100) which is the biggest production and processing facility of Mellitah oil and Gas B.V Company. It is located in the middle desert of Libya, approximately 400 km South of Benghazi and about 60 km South East of Jalo Oasis. According to information provided in the year 2010, the field produced about eighty-five thousand barrels of crude oil per day. Sixteen soil samples named S_1 to S_{16} have been randomly collected from several points of the evaporation ponds. For the control samples four of them C_1-C_4 were collected far from the ponds, about 1000 m each from North, South, East and West. All of the samples were collected in line with cited recommendations. There are five ponds of different sizes which are about 1 km far south of the field. They have sloping edge but no lining, with a 217.776 m^3 total capacity, soil sampling points are as shown in Fig. 1.

Gamma-ray spectrometry: The detector used in this study is a high-resolution, low-background gamma-ray spectrometer based on a HPGe detector having efficiency of 34% from ORTEC, housed in the Nuclear Radiation Laboratory (NRL) at nuclear engineering department, Tripoli University. The energy resolution, expressed in terms of its FWHM was 1.8 keV at the 1332.5 keV^{60}Co transition. To attenuate cosmic and other outside radiation, a lead castle of approximately 10 cm thick with a copper lining on the inside to absorb any lead X-rays that are produced, shields the detector. To optimize the detection efficiency in measuring environmental samples, a large quantity of the sample must be as close to the

Fig. 1: Soil sampling points

detector crystal as possible. In order to achieve this Marenelli beakers were used. The Marenelli beaker used in this study is a one litre polypropylene beaker with a 90 mm annular bottom. The beaker slides over the HPGe detector. When γ-ray spectrometry is used for the measurement of natural radioactivity in environmental soil and sand samples, the samples must be properly sealed for about four weeks to obtain radioactive secular equilibrium between ^{222}Rn (radioactive noble gas), its decay products (^{214}Pb and ^{214}Bi) and radium (^{226}Ra), from the ^{238}U decay series. In the ^{232}Th decay series the radon isotope (^{220}Rn) poses no serious problem because of its short half-life of 55 seconds and in the ^{40}K decay series no equilibrium is needed (Van, 1994). Efficiency calibration for the specified geometries and energies was performed using a cocktail standard source as a reference standard. In this survey, the amount of three radionuclides ^{226}Ra, ^{232}Th and ^{40}K present in the samples was evaluated. Obviously radium concentration was considered more important than other elements due to its higher relative solubility in wastes, oil and gas production equipment (Khodashenas *et al*., 2012).

NORM regulations and guidance: Regulation of NORM presents a range of new challenges for both regulators and operators. Libya has no national regulations specific to NORM, however, general radiation protection regulations were issued in 1982. There are many international NORM regulations for sludge and scale as for example Louisiana regulations; Georgia regulations; Texas regulations]; IAEA while the Saudi Aramco regulations for NORM included regulations for NORM soil contamination (Cowie *et al*., 2012); Syrian regulations (Al-Masri and Suman, 2003) and the Norway regulations for clearance and disposal of NORM.

There is also guidance provided for the purpose of limiting NORM contamination such as oil industry international exploration and production forum guidance; Canadian guidelines for the management of naturally occurring radioactive materials (Act, 2000). There is also the European standard, council of the European union.

The regulations adapted by the Saudi Aramco are based on best prevailing guidance and practices throughout the oil and gas industry. Since, this study is only focusing on soil contamination with NORM the following limits are provided by Saudi Aramco:

- Ra-226 soil contamination should not exceed 0.185 Bq g^{-1} (5 pCi g^{-1})
- U-238 soil contamination should not exceed 3.3 Bq g^{-1} (90 pCi g^{-1})

The Syrian regulations are mainly criteria for disposal and clean-up of contaminated soil have been defined as follows:

Table 1: The regulation of exemption and disposal levels of Ra-226 in different countries

Countries	Ra-226 exemption level (Bq kg^{-1})	Ra-226 disposal level (Bq kg^{-1})
Saudi Aramco	<185	-
Syria	<150	>5200
Norway	<500	>5000

- Soil containing not >0.15 Bq g^{-1} of ^{226}Ra does not need any treatment
- Soil having specific activity of ^{226}Ra higher than 5.2 Bq g^{-1} need to be managed as radioactive waste
- Contaminated areas containing ^{226}Ra with concentration between 0.15 and 5.2 Bq g^{-1} need a special treatment on site to reduce the exposure to a value below 100 µSv/a

The Norway regulation for clearance and disposal of NORM waste has been classified only according to the repository level, meaning that this level has also been used as a clearance level. The repository level is 5 Bq g^{-1} for ^{226}Ra, ^{228}Ra and ^{210}Pb. With the implementation of the proposed clearance level of 0.5 Bq g^{-1}, the waste industry is currently working towards treating waste from the petroleum industry with activity between the clearance level and repository level as NORM waste and adapting the methods for its handling and disposal accordingly. In Sweden when the activity concentrations of ^{238}U and ^{232}Th decay series radionuclides in NORM waste are above 1 Bq g^{-1} but do not exceed 10 Bq g^{-1}, the waste can behandled and disposed of according to the guidelines in regulation SSMFS 2011 (Koufakis, 2013).

It is clear that the existing regulations in the above mentioned countries are more or less the same. To make it clear the following Table 1 for the different country limits is presented.

In this study these limits are applied to evaluate the management strategy for ponds NORM contamination in the south eastern oilfield, in the Libyan desert For the lower limit for exemption is 150 Bq kg^{-1} while the lower limit for disposal is 5000 Bq kg^{-1}.

RESULTS AND DISCUSSION

NORM (contaminated soil) in the evaporation ponds: The content of natural radionuclides in soil of Libya has been reported by UNSCEAR (2008) global survey on exposures to natural radiation sources. The concentration ranges in soil for ^{40}K, ^{238}U, ^{226}Ra and ^{232}Th are 265-282, 8.7-12.8, 8.3-9.4 and 7.6-9.7 Bq kg^{-1}, respectively. Soil samples of the evaporation ponds S_1–S_{16} show higher radio-activities of ^{226}Ra and ^{232}Th than those in control samples C_1–C_4 as illustrated in Fig. 2-4. The highest average radioactivity of ^{226}Ra was 1000 Bq kg^{-1} in sample S_{14}; while the lowest radioactivity was 83 Bq kg^{-1} in sample S_9. Figure 3 shows that the highest radioactivity of ^{232}Th was 315 Bq kg^{-1} in sample S_{16} while the lowest average radioactivity was 59 Bq kg^{-1} in sample S_{13}. The

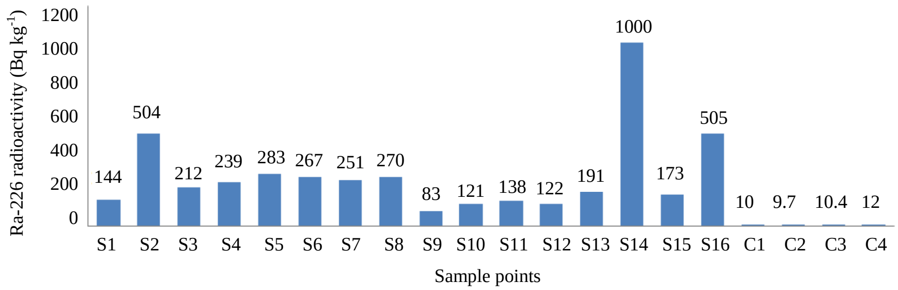

Fig. 2: Ra-226 radioactivity in the oilfield ponds (A/100)

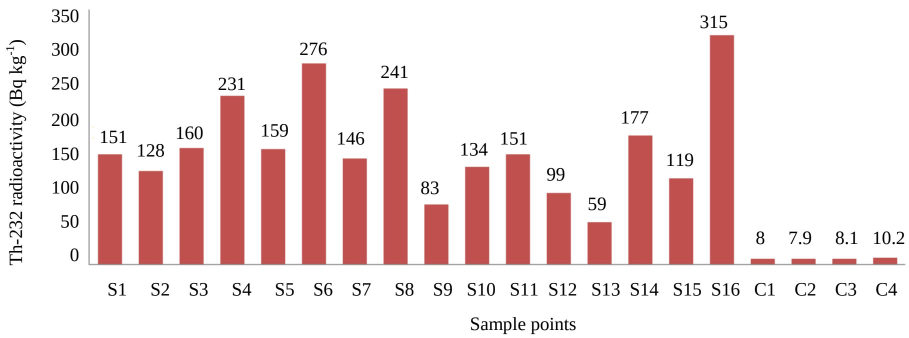

Fig. 3: Th-232 radioactivity in the oilfield ponds (A/100)

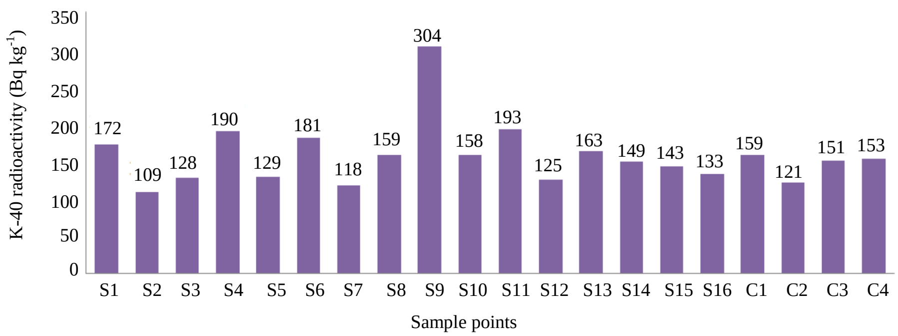

Fig. 4: K-40 radioactivity in the oilfield ponds (A/100)

highest radioactivity of ^{40}K was 304 Bq kg^{-1} in sample S_9; while the lowest radioactivity was 109 Bq kg^{-1} in sample S_2 as shown in Fig. 4.

According to the above suggested limits for soil NORM contaminations presented in Table 1, it can be seen that eleven samples of the soil samples presented in Fig. 2-4 are higher than the exemption limit of 150 Bq kq^{-1} of Ra-226 and no sample exceeds the disposal limit of 5000 Bq kg^{-1}. This means according to and the Libyan suggested level of regulations there is a need to reduce the concentrations of NORM to acceptable levels below 150 Bq kg^{-1}. This can be done by special treatment on site.

Radiation hazard assessment: In addition to the effective dose, a number of parameters were also used for the evaluation of the potential hazard associated with the natural radionuclides. These include radium equivalent activity and the external and internal hazard indices.

Radium equivalent activity: The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in materials can be compared by using the concept of radium equivalent activity ($^{226}Raeq$), which is a common radiological index used to evaluate the actual radioactivity in the materials by a single quantity. This takes into account the associated hazards. The Ra_{eq} of the activities of natural radionuclides in individual samples was calculated by using formula

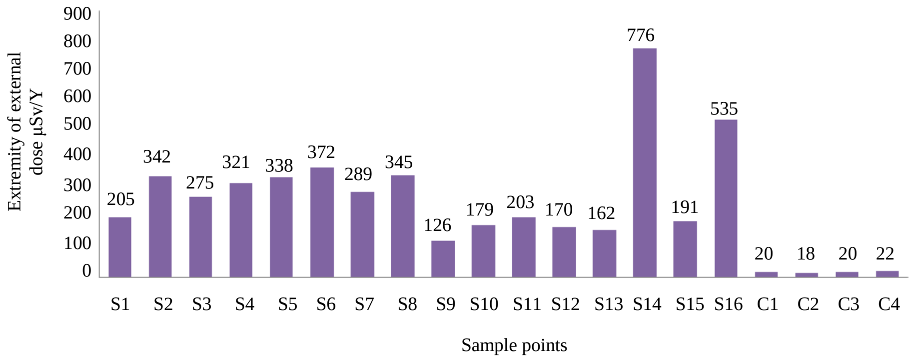

Fig. 5: External dose in the sample area of the oilfield ponds (A/100)

(Beretka and Mathew, 1985). $Ra_{eq} = {}_{Cra}+1.43C_{Th}+0.077C_K$ where C_{Ra}, C_{Th} and C_K are the activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

External dose: Analyses on extremity of the external dose on oilfield ponds (A/100) samples indicate they are much higher the average background. Results are shown on Fig. 5.

CONCLUSION

This investigation is carried out on a small and limited area and it is known that NORM concentrations vary because of the geological characterisation of different oilfields and it could be concluded that:

To prevent underground water contamination and reduce soil contamination ponds lining should be implemented for new pits.

More samples are needed to evaluate the level of contaminations in this oilfield. NORM in Scale and sludge in this oilfield need to be analysed in a new future study.

There was a need to regulate and reduce the radiological risk arising from TENORM to as low as reasonably achievable taking into account the economic and political factors of the country. It is suggested that the Libyan regulatory office adapts the Ra-226 exemption level of <150 Bq kg^{-1} and the disposal level of >5000 Bq kg^{-1} for soil contamination.

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