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Comparison Between, NORM (Naturally Occurring Radioactive Materials) of, Agricultural Soil Sample (tomato field treated with phosphate fertilizer) Relatively Close to an Oil Field, and Wastes Samples (scale and sludge) of the Same Oil Field.

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ABSTRACT: *This paper is an overview comparison of NORM (Naturally Occurring Radioactive Materials). Soil sample was collected from a tomato field which was treated by phosphate fertilizers, and scale and sludge samples were collected from an oil field. The two fields are relatively close (less than 60 km). The results were as follows:*

Table 1

	^{40}K Potassium activity concentration (Bq/kg)	^{238}U Series Uranium activity concentration (Bq/kg)	^{232}Th Series Thorium activity concentration (Bq/kg)	Ra_{eq} Radium equivalent activity (Bq/kg)
Soil samples	1040.43± 8.32	2458.48± 19.66	336.47± 2.68	2951.62± 23.6
Scale sample	844.21± 2.53	3681.56± 29.44	33.04± 0.26	3728.81± 29.8
Sludge sample	104.56± 0.83	42.81± 0.33	26.71± 0.20	104.32± 0.66

Table 2

	D Absorbed dose rate in air (nGy/h)	AEDE Annual effective dose equivalent (mSv/y)	H_{ext} External hazard index	H_{int} Internal hazard index
Soil samples	1345.54±10 .76	1.65±0.012	7.97±0.06	14.62±0.08
Scale sample	1720.84±13 .76	2.11±0.016	10.07±0.07	20.02±0.09
Sludge sample	48.53±0.38	0.059±0.03	0.39±0.02	0.28±0.001

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INTRODUCTION

The most common radiation sources to which humans are exposed arise from radionuclides in the earth's surroundings and the interaction of cosmic rays on the earth's atmosphere [1]. This exposure to naturally occurring radiation accounts for up to 85% of annual exposure dose received by the world population. The majority of naturally occurring radionuclides (NORs) belong to the decays in the ^{235,238}U and ^{232}Th series and the single decay radionuclide, ^{40}K [2].

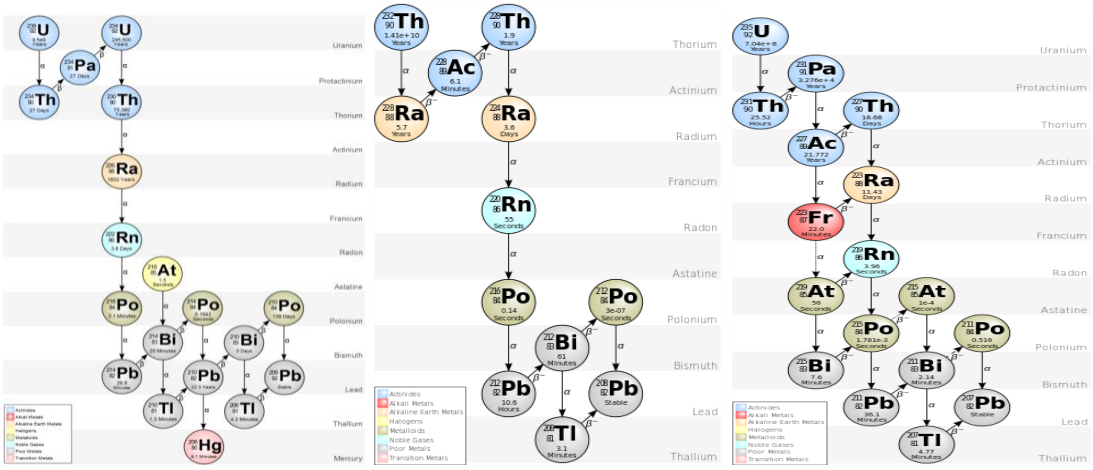

Fig-1 The natural radioactive decay series [3]

In the oil and gas industry: NORs are present at varying concentrations in the Earth's crust and consequently also will be present in natural concentrations in gas and oil reservoirs. The NORs concentrations in well fluids may become enhanced due to extraction processes and subsequently form NORs enriched deposits within production facilities thereby forming NORM (Naturally Occurring Radioactive Materials). Examples include produced water, scales, sludge and pigging debris. Uncontrolled work activities involving NORM can lead to unwanted exposure and dispersal posing a risk to human health and the environment. [4].

Phosphate processing. This industry may be subdivided into (a)wet processing, (b) thermal processing, and (c) fertilizer production. The primary product is phosphoric acid. In the thermal process, the product may be phosphorus or, using nitric acid, phosphoric acid. Phosphoric acid is used in the manufacture of fertilizers. In the wet phosphate processing industry, phosphogypsum is produced as a by-product. The thermal process (using cokes and silica) produces a slag (CaSiO2) as a waste product.

In the agriculture field: It is important to measure natural radioactivity, not only in the phosphate rock, but also in different types of fertilizers and by-products, because the high radioactive content may lead to significant exposure of miners, manufacturers and end users. The fertilizers play a vital role in agriculture, this led to the spread of the industry of fertilizers all over the world widely and also to the extraction of raw materials of phosphate which are appearing on the surface of the earth[5]. Phosphate is the main material used for the fertilizers**,** so NORM resulting from phosphate of fertilizers is similar to the oil industry NORM**.**

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In this research, as a result of the permanent complaint of population of the areas near to the oil fields of the environmental impact of these oil fields residues on their agricultural environment and their health , a comparison was made between the NORM of agricultural phosphate used in the tomato farms and the NORM resulting from the neighboring oil fields, The risk assessment indicators, mentioned to the two NORMs radiation activity were very close, this truth is not clear for the **population** of these areas.

MATERIAL AND METHODS

After the routine preparation of the samples, as cleaning, drying and filtering, as well as calibration of the detector, the samples activity concentration were measured using a hyper-pure germanium (HPGe) detector, this detector with efficiency 40%, vertical position and resolution 1.89 kev is present in the protection building in the Center of Nuclear Research (CNR)- Tajoura (Tripoli-Libya). Figure 2 shows the study area and a summary of the initial information about samples is given in table 3.

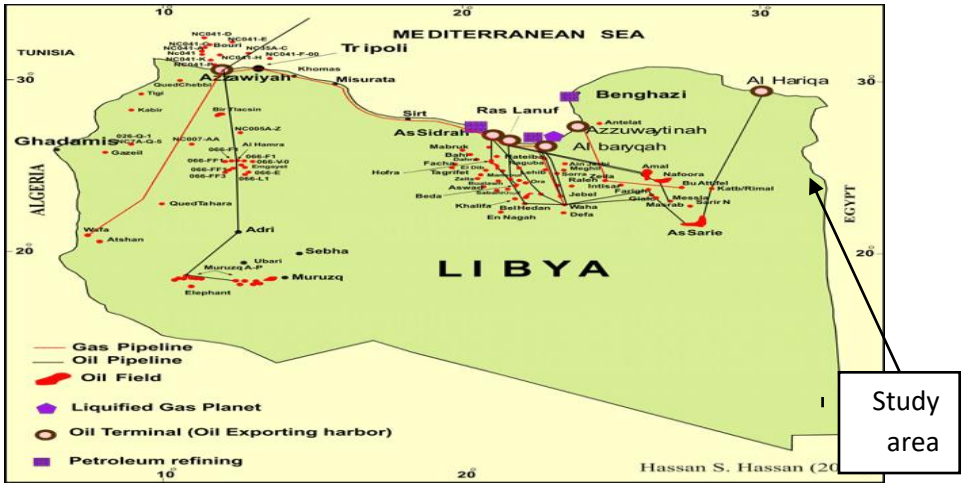

Fig 2- The Location of the Study Area in the Libyan Map Oil

Table 3- The Initial Information about Samples

The sample	Net Weight (kg)	Collection Date	Detection Date	Duration of Detection
Soil	0.9	March 2018	May 2018	48 Hour
Scale	0.7	April 2018	May 2018	48 Hour
Sludge	0.9	April 2018	May 2018	48 Hour

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Activity concentration determination

Each prepared sample was measured for 48 hours. The net counts under the full-energy peak areas, the counting time, the absolute full-energy peak efficiency for the energy of interest and the gamma-ray emission probability corresponding to the peak energy were used to determine the activity concentration of a particular radionuclide in the measured samples. Prior to that a background radiation level of the HPGe system was obtained by counting deionized water for the same duration (two days) in a 550 ml Marinelli beaker which

again had the same geometry as that in counting of the sample. The background spectrum was later subtracted from each sample spectra before activity concentration determination. The quoted specific activity concentration values (in Bq/kg) assume secular equilibrium for the different isotopic activities in the decay chains and is defined as the activity per unit mass of the sample [6], expressed as:

$$A = \frac{C_n}{Eff\ \ I_\gamma W} \qquad \rightarrow (1)$$

where A is the activity concentration of a particular nuclide in units of Bq/ kg, C_n the net count (background subtracted) of the corresponding full energy peak($net\ CPS\ samples$), Eff the absolute full-energy peak detection efficiency, I_γ the emission probability per decay corresponding to the specific gamma-ray energy, ts the counting time in second and W the mass of the soil sample in kg.

Radiological risk assessment

One of the main objectives of the radioactivity measurement in environmental samples is to estimate the radiation exposure dose and to assess the biological effects on humans. The assessment of radiological risk can be considered in various terms.

The dose rates (D) for the measured samples were determined from the specific activity concentration, in addition to the associated radiological risks from the absorbed dose at 1 m above the ground surface, viz, the radium equivalent activity (Raeq) which is used to assess hazards associated with materials containing ^{226}Ra, ^{232}Th and ^{40}K nuclides; internal hazards index (H_{int}), external hazard index (H_{ext}) and finally the annual effective dose equivalent (AEDE) were calculated using the following references[7]. Hint gives a measure of internal exposure due to radon (^{222}Rn) and the index must be less than unity to be within the safety threshold, whereas the H_{ext} index evaluates external radiation exposure from radium containing materials. A value of Hext =1 represents the maximum permissible value equivalent to a radium equivalent activity of 370 Bq/kg.

$Ra_{eq} = A_u + (A_{Th} \times 1.43) + (A_K \times 0.077) \rightarrow$ (2)

$D = (0.462 \times A_U) + (0.604 \times A_{Th}) + (0.0417 \times A_K) \rightarrow (3)$

$AEDE = D \times 1.23 \times 10^{-3}$ mSv/y $\rightarrow (4)$

$$H_{int} = \left(\frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \leq 1 \qquad \rightarrow (5)$$

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$$H_{ext} = \left(\frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \leq 1 \quad \rightarrow (6)$$

The units for R_{aeq}, D and AEDE are given in Bq/kg, nG/hr, and mSv/year respectively. The symbols A_U, A_{Th}, and A_K represent the specific activities in (Bq/kg) for the ^{238}U chain, the ^{232}Th chain and ^{40}K respectively.

RESULTS AND DISCUSSION

The activity concentration

The activity concentration for each 3 samples (soil, scale and sludge) is shown in Fig. 2. Results are for the ^{238}U, ^{232}Th primordial decay chains, in addition to the singly decaying ^{40}K. It can be observed that the same activity level between soil and scale samples, and very low value as expected of sludge sample.

The values obtained due to ^{238}U, ^{232}Th, and ^{40}K for soil sample was 2458.48, 336.47 and 1040.43 Bq/kg, scale sample was 3681.56, 33.04 and 844.21 Bq/kg and sludge sample was 42.81, 26.71 and 104.56Bq/kg respectively.

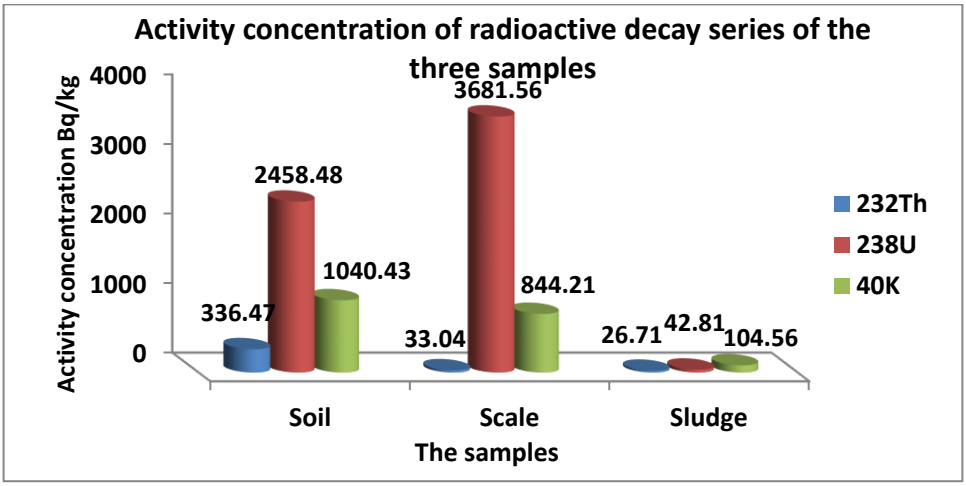

Figure 2

The hazards indicators

The dose rates (D) and radium equivalent activity (Ra_{eq}) for each sample are shown in Fig 3, as a result of activity concentration the same direction is clear in these indicators, it means high values for soil and scale samples and low values for sludge samples. The values obtained of D and Ra_{eq} for soil sample was 1345.54nGy/h and 2951 Bq/kg, scale sample was 1720.84nGy/h and 3728.81 Bq/kg and sludge sample was 104.32nGy/h and 48.53Bq/kg respectively.

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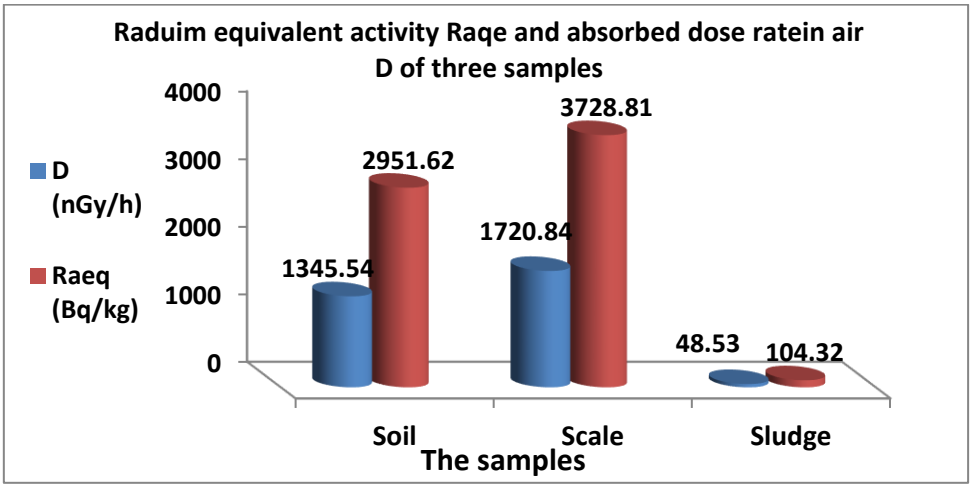

Figure 3

Internal hazards index (H_{int}), external hazard index (H_{ext}) and the annual effective dose equivalent (AEDE) for each sample are shown in Fig 3**.** The values obtained of H_{in}, H_{ext} and AEDE for soil sample was 14.62, 7.97 and 1.65mSv/y, scale sample was 20.02, 10.07 and 2.11mSv/y and sludge sample was 0.28, 0.39 and 0.059mSv/y respectively.

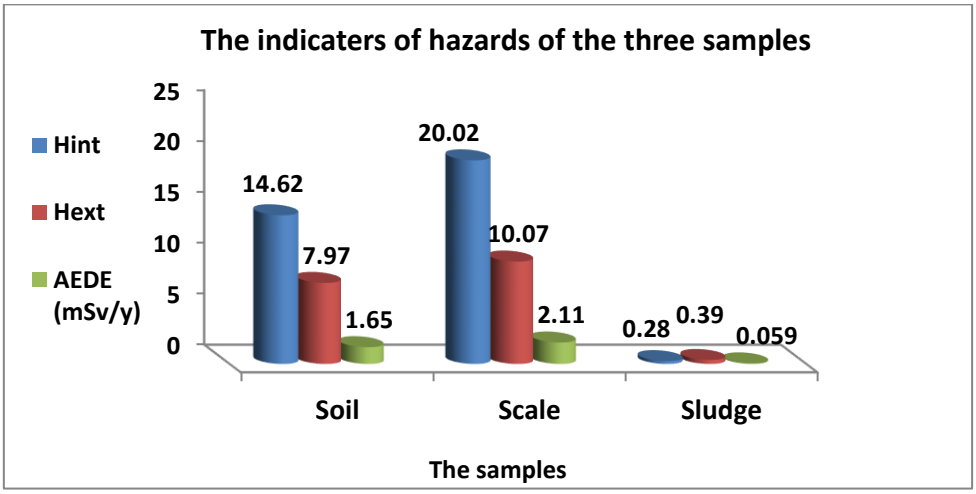

Figure 4

The ICRP60,1990 recommends that any exposure above natural background radiation should be keep it as low as reasonably achievable (ALARA principle) but below the individual dose limits, which for radiation workers averaged over 5 years is 100mSv and for members of the general is 1mSv/y. These dose limits have been established on the prudent approach by assuming that there is threshold dose below for which there would be no effect. This means that any additional dose will cause a proportional increase in the chance of the health effect.

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The activity concentrations and evaluated doses from this study are shown in Tables 4 and 5 compared to exemption level and dose limit of general Public of Annual Effective Dose Equivalent(AEDE).

The AEDE is the most important hazard indicator, which indicates the effective dose which may be the human exposure to annually, the values of AEDE of soil and scale samples in table 1 equal approximately 1.5 and 2 times of the dose limit of the general public. Of course these values calculated depend on the concentration activity of ^{238}U(^{226}Ra), ^{232}Th and ^{40}K of 3 samples, and these values are absolutely true, but in the same time should be observed that the harmful effect of these doses is more complicated than it seems, because there are many considerations must be taken in account, like the real time of exposure, the distance of exposure and the energy of radiation. It means that the real exposure is less than these values, maybe two or three times more than what was calculated.

Table 4- The exemption level compared to the current work

The Sample	**Level of Current work (Bq/kg)**			**Exemption Level (Bq/kg)**			**Reference**
	^{238}U	^{232}Th	^{226}Ra	^{238}U	^{232}Th	^{226}Ra	
Soil	2458.48	1040.43	2951.62	5500	1100	1100	4
Scale	3681.56	844.21	3728				
Sludge	42.81	26.71	104.32				

Table 5 The Annual Effective Dose Equivalent (AEDE) of 3 samples

	Soil sample	**Scale sample**	**Sludge sample**	**Dose limit of general Public**	**Reference**
AEDE Annual Effective Dose Equivalent (mSv/y)	**1.69**	**2.11**	**0.059**	**1**	4

A summary is given in Table 6 of the activity concentrations for the phosphate fertilizers sample compared to other reported values from across the Middle East and some other countries. This comparison illustrated that the magnitude of all activity concentrations of this study are high which is comparable to the other countries, maybe except the level of ^{226}Ra in Sudan.

The danger posed by phosphate fertilizer is its presence in the midst of residential communities, the great potential for its infiltration into the groundwater and its presence in the human food chain. In contrast, all these risks are less likely for the oil fields wastes, especially if dealt with professionally and carefully.

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Table 6- Comparison between activity concentration of 226Ra 232Th and 40K in Bq/kg soil phosphate of present work with that of other countries

Country	^{226}Ra	^{232}Th	^{40}K	Reference
Present work	**2951**	**336**	**1040**	
Egypt	**839**	**394**	**129**	**8**
Tunisia	**821**	**29**	**32**	
Algeria	**619**	**64**	**22**	
Morocco	**1600**	**20**	**10**	
Jordan	**1044**	**2**	**8**	**9**
Sudan	**4131**	**7.5**	**62**	
Finland	**10**	**10**	**110**	
USA (west)	**1600**	**20**	**N.F**	
USA (East	**1000**	**20**	**N.F**	

CONCLUSIONS

The purpose of this research is to try to raise awareness and to send a message to the population of areas close to the oil fields, that the radioactive contamination they hear about it which comes from the oil fields near to their residential communities, farms and pastures is not as they believe in such danger, for example materials and fertilizers using in their farms, contain natural radioactive materials sometimes exceeding what reached them from neighboring oil fields.

To clarify it more, the largest releases of radionuclides to water come from the phosphate processing, followed by oil and gas production and primary iron and steel production. As an example, two phosphoric acid plants in the Netherlands are responsible for some 90% of all discharges of ^{210}Pb and ^{210}Po to water. These two plants release about 0.6 - 0.8 TBq of ^{226}Ra per year, which is comparable to the estimated annual release of ^{226}Ra with processed water into the North Sea by the offshore oil production industry in the United Kingdom, Norway, the Netherlands, and Denmark[9] .

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