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Measurement of Activity Concentration, Absorbed Dose Rate and Annual Effective Dose of Natural Occurring Radioactive Material (NORM) In Samples Encountered During Oil & Gas Industry

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Abstract: *Uncontrolled activities associated with enhanced levels of NORM, can contaminate environment and pose a risk to human health. These risks can be alleviated by the adoption of controls to identify where NORM is present, its concentration and control NORM-contaminated equipment and waste. This paper try to identify type, activity concentration, absorbed dose rate and annual effective dose of radionuclides present in samples collected from oil fields in West Kordofan state which include Heglig, Bamboo, Diffra and Neem oil fields.*

I. Introduction

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 recovered. During the production process, NORM flows with the oil, gas and water mixture and accumulates in scale, sludge and scrapings. The level of NORM accumulation can vary substantially from one facility to another depending on geological formation, operational and other factors. [1]
The formation water contains Group II (Periodic Table) cations of calcium, strontium, barium and radium dissolved from the reservoir rock. As a consequence, formation water contains the radium isotopes ^{226}Ra from the ^{238}U series and ^{228}Ra and ^{224}Ra from the ^{232}Th series. All three radium isotopes, but not their parents, thus appear in the water co-produced with the oil or gas. [2]

II. Research Methodology

2.1 Fields trips

Fields trips were carried out to four oil fields in West Kordofan State (Blocks 2 & 4). The trips include "Heglig, Neem, Diffra and Bamboo oil fields", where Heglig & Bamboo located in block "2", Neem & Diffra in block "4" (Fig.1). The aim of these trips was to collect samples for analysis and so to determined type, activity concentration of Natural Occurrence Radioactive material, absorbed dose rate and annual effective dose from radionuclides in these samples. Kind of samples and photos of some sample sources are in Appendix.

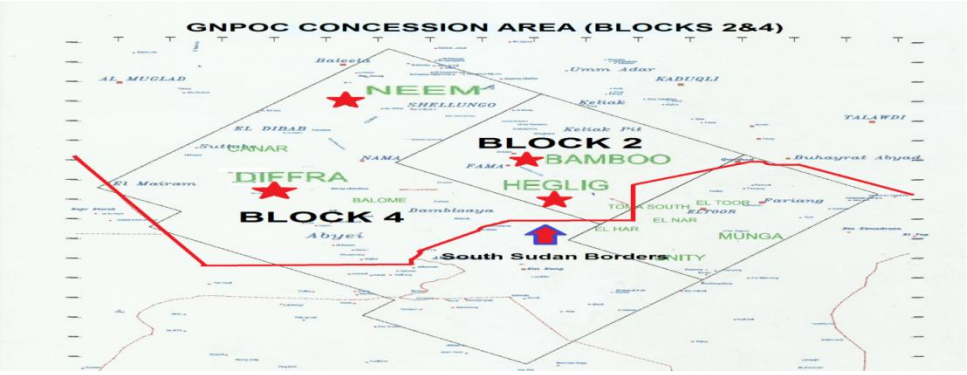

Figure 1: Map showing Sampling areas

2.2 Sampling methods and analysis preparations

Samples were taken manually after full PPE (Disposable coverall, respiratory mask, gloves, glass, helmet and safety boot). Sludge samples were taken with plastic shovel, sand with plastic shovel and brush, scale samples were taken by using hammer, chisel and brush.Each sample was contained in plastic bags. The samples was weighted and placed into Marinelli beakers and stored for more than four weeks before counting, in order to allow of secular equilibrium of parent with its short-lived progeny to take place. The samples have been characterized using gamma spectroscopy through using of thallium activated sodium iodide NaI (TI) spectrometer.Calibration process carried out for gamma spectroscopy using standard calibration source MW652 as reference source. Each sample was placed onto the detector and measured for at least three hours. The ^{238}U concentration was determined from the average concentrations of the ^{214}Bi (609 Kev),^{214}Pb (352 keV) and ^{214}Pb (295 keV) decay products. ^{232}Th concentration was determined from the concentrations of the ^{212}Pb (238 keV). ^{40}K concentration was determined directly.

2.3 Energy calibration

Energy calibration sources were performed using cesium-137 or cobalt-60. Because the channel number is proportional to energy, the channel scale can then be converted to an energy scale (Fig. 2).

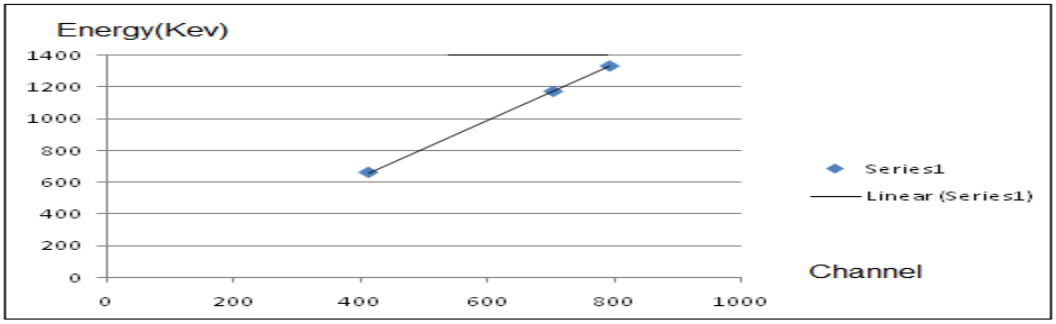

Figure 2: Energy calibration carve

2.4 Efficiency calibration

Detector efficiency calibration was performed using a mixed radionuclide sources (MW652) in Marinelli beaker geometry. The following equation (Eq. 2.1) was used to obtain the efficiency curve of the detector for different energies:

$$\eta = \frac{Count \quad (CPS)}{I_\gamma \times A \ (Bq \ / \ Kg)} \qquad (2.1)$$

Where, η is the efficiency of the detector at specific energy; I_γ Iγ is gamma intensity, and A is the activity of the standard.

Table 2.1 illustrate Radionuclides, energies and corresponding efficiencies of the radionuclides in the standard calibration source.

Table 2.1: Efficiency for calibration source

Radionuclide	Energy (KeV)	CPS	Activity	i_γ	Efficiency
Cs - 137	662	27.87	2147.005	0.851	0.015254
Co - 60	1173	5.22	805.5982	0.9997	0.006482
Co - 60	1333	4.74	805.5982	0.9998	0.005885

III. Calculations

- The activity concentrations were calculated for the radionuclides in the samples using the following equation (Eq. 3.1)

$$A = \frac{N}{I_\gamma \times \eta \times m} \qquad (3.1)$$

Where:

"A" : is activity concentration of the sample (Bq/Kg).
"N" : is the net area of the peak (count per second).
"I_γ" : is gamma intensity.
"ŋ" : is the efficiency of the detector at specific energy.
"M" : is the mass of the measured sample (kg).

- The gamma dose rate (D) in nGy/hr caused by naturally occurring radioactive materials in air at 1m above the ground surface can be estimated using the following equation (Eq. 4.2) .[3]

$$D = 0.462A_{U-238} + 0.604A_{Th-232} + 0.0417A_{K-40} \quad (3.2)$$

Where:
A_{U-238}: Activity concentration of ^{238}U in (Bq/Kg).
A_{Th-232}: Activity concentration of ^{232}Th in (Bq/Kg).
A_{K-40}: Activity concentration of ^{40}K in (Bq/Kg).

- The annual estimated average effective dose equivalent received was calculated using a conversion factor of 0.7 $SvGy^{-1}$. The outdoor occupancy factor is about 0.2. The annual effective dose (AED) is given by the following equation[4].

$$AED\ (mSv/y) = D\ (nGy/h) \times 8760\ (h/y) \times 0.2 \times 0.7(Sv/Gy)10^{-6} \quad (3.3)$$

IV. Result and discussion

The results of samples activity concentrations (Bq/Kg), absorbed dose rate (nGy/h) and annual effective does (mSv/y) were tabulated in Tables 4.1, 4.2, 4.3 and 4.4.

Table 4 .1: Neem Field samples activity concentration, absorbed dose rate and annual effective dose results:

No.	Sample ID	Concentration of ^{238}U (Bq/Kg)	Concentration of ^{232}Th (Bq/Kg)	Concentration of ^{40}K (Bq/Kg)	Absorber Dose Rate (nGy/h)	Annual Effective Dose (mSv/y)	Kind of sample
1	NHT1	4106.9	2736.7	3397	3692.01	4.53	Scale
2	NPT1	944.8	1156	9294	1522.28	1.87	Scale
3	NFT1	663.5	595.1	614.89	691.62	0.85	Sand
4	NCI2	56.75	91.7	353.52	96.35	0.12	Sludge
5	NHTD1-2	2183.84	1237.81	112.48	1761.26	2.2	Scale
6	NPTD2-2	1323.49	1203.80	16.06	1339.22	1.64	Sand
7	NHD3-2	1430.00	1081.05	64.28	1316.29	1.61	Sand
8	NUND4-2	2807.36	3466.24	111.25	3395.25	4.16	Sand
9	NHTD5-2	2641.77	2517.52	37.4	2742.64	3.36	Sand
10	NPTD6-2	2592.78	3137.52	110.28	3097.53	3.8	Sand
11	NS-28	743.15	733.52	16.07	787.05	0.97	Scale
12	NS-29	1392.74	1373.87	1285.53	1526.87	1.87	Scale
13	NS-30	455.54	320.91	16.07	404.96	0.5	Sludge
14	NS-31	655.36	396.34	16.07	542.84	0.7	Sludge
15	NS-32	647.81	393.38	16.07	537.56	0.66	Sludge

Table 4. 2: Diffra Field samples activity concentration, absorbed dose rate and annual effective dose results:

No.	Sample ID	Concentration of ^{238}U (Bq/Kg)	Concentration of ^{232}Th (Bq/Kg)	Concentration of ^{40}K (Bq/Kg)	Absorber Dose Rate (nGy/h)	Annual Effective Dose (mSv/y)	Kind of sample
1	DHA-1	603.43	385	504.65	532.37	0.65	Scale
2	DO-1	9.7	16.19	92.62	18.12	0.02	Crude Oil
3	DP-1	64.11	63.69	22.13	69.01	0.08	Sludge
4	DW-2	358.05	375.88	133.9	398.03	0.49	Scale
5	DPR-2	81.21	90.21	96.41	96.03	0.11	Sludge

Table 4.3: Bamboo Field samples activity concentration, absorbed dose rate and annual effective dose results:

No.	Sample ID	Concentration of ^{238}U (Bq/Kg)	Concentration of ^{232}Th (Bq/Kg)	Concentration of ^{40}K (Bq/Kg)	Absorber Dose Rate (nGy/h)	Annual Effective Dose (mSv/y)	Kind of sample
1	BST-i2	13.89	14.6	196.28	23.42	0.02	Sand
2	BST-ii2	14.998	25.16	163.2	28.93	0.04	Sand
3	BTP-1	71.36	52.59	65.6	67.47	0.08	Sand
4	BNP-1	67.63	68.92	62.16	75.46	0.09	Sand

Table 4.4: Heglig Field samples activity concentration, absorbed dose and annual effective dose results:

No.	Sample ID	Concentration of ^{238}U (Bq/Kg)	Concentration of ^{232}Th (Bq/Kg)	Concentration of ^{40}K (Bq/Kg)	Absorber Dose Rate (nGy/h)	Annual Effective Dose (mSv/y)	Kind of sample
1	HPR-1	23.3	16.19	238.65	30.49	0.03	Sludge
2	HST-1	75.6	77.5	169.6	88.81	0.11	Sludge
3	HG-1	59.13	52.42	224.7	68.35	0.08	Contaminated grass
4	HSD-1	319.41	308.58	145.5	340.02	0.42	Sand
5	HSD-2	186.27	257.32	16.07	242.15	0.3	Sludge
6	H56-2	306.30	436.27	80.35	408.37	0.5	Sand
7	HSTD-2	303.62	32.54	32.14	161.27	0.2	Sludge
8	HFT-2	1961.64	2430.63	45.91	2376.29	2.9	Sand

The results of samples analysis indicate existence of elevated radionuclides concentration of samples collected from block "4" than samples collected from block "2" (This is due to absence of scale which had high activity concentration from block 2). It is recommended that the acceptable total absorbed dose rate by the workers in areas containing γ-radiations from ^{238}U and ^{232}Th series and their respective decay progenies, as well as ^{40}K, must not exceed 55 nGy/h[5]. The International Commission on Radiological Protection (ICRP) has recommended the annual effective dose equivalent limit of 1 mSvy^{-1} for the individual members of the public [6] and incidentally exposed workers [7] and 20 mSvy^{-1} for the radiation workers [6](Occupationally exposed worker)[7].

Where:

Members of the Public are persons who have no occupational exposure to NORM.

Incidentally Exposed Workers are employees whose regular duties do not include exposure to NORM sources of radiation. They are considered as members of the public who work in an occupational exposure environment.

Occupationally Exposed Workers are employees who are exposed to NORM sources of radiation as a result of their regular duties. They are classified as NORM workers working in an occupational exposure environment [7].

Figure 4 show Comparison of average activity concentration of radionuclides found in Scale, sludge and sand. Figures 5,6,7 and 8 illustrates comparison between Annual Effective Dose (AED) result from different fields samples, and recommended AED for members of the public/incidental worker and occupationally workers.

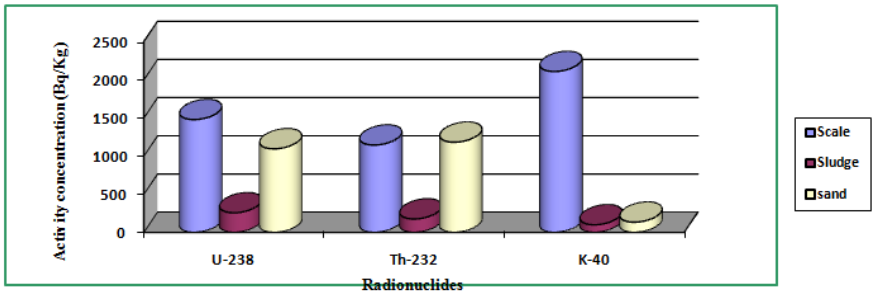

Figure 4: Comparison of average activity concentration of radionuclides found in Scale, sludge and sand

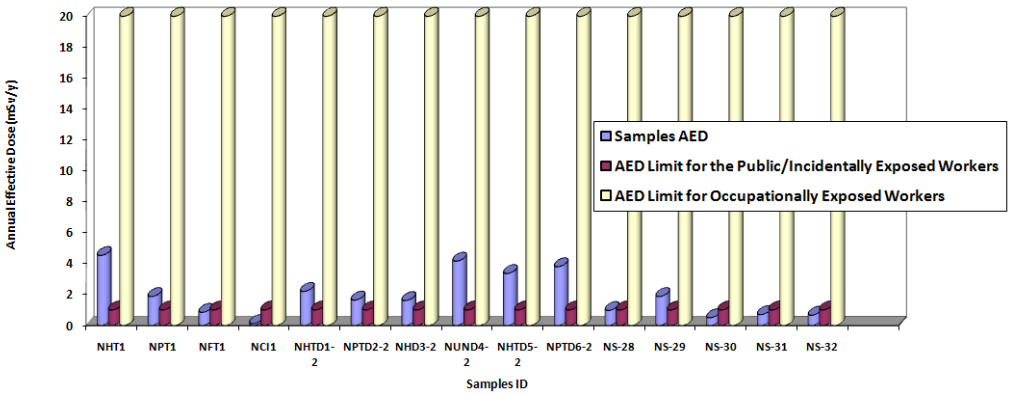

Figure 5: Comparison between Annual Effective Dose (AED) result from samples and AED limits for members of the public/Incidentally exposed workers and occupationally exposed workers - (Neem Field)

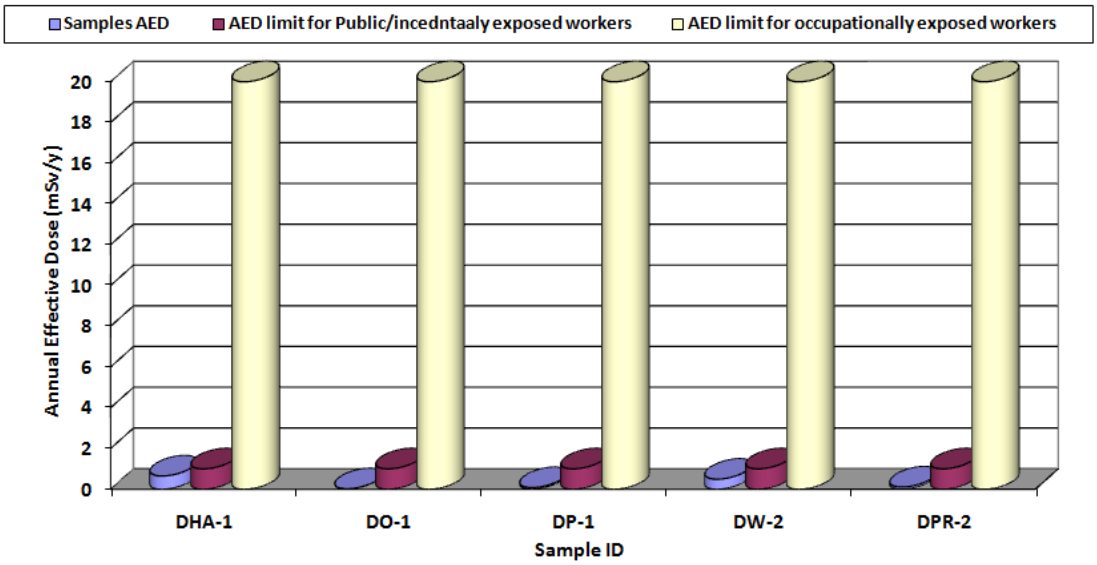

Figure 6: Comparison between Annual Effective Dose (AED) result from samples and AED limits for members of the public/Incidentally exposed workers and occupationally exposed workers - (Diffra Field)

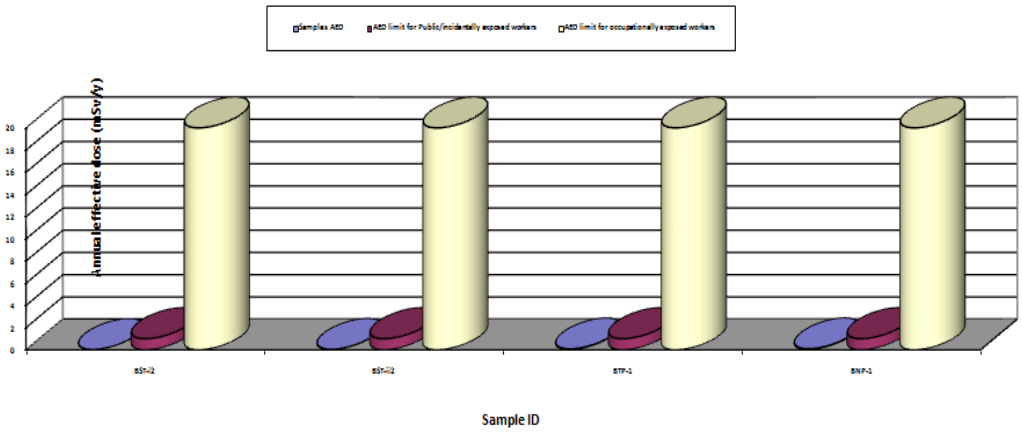

Figure 7: Comparison between Annual Effective Dose (AED) result from samples and AED limits for members of the public/Incidentally exposed workers and occupationally exposed workers - (Bamboo Field)

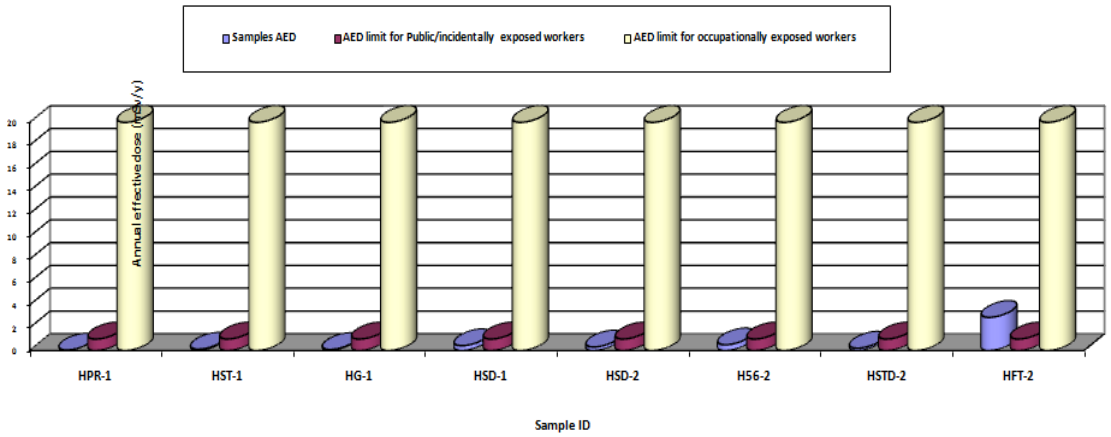

Figure 8: Comparison between Annual Effective Dose (AED) result from samples and AED limits for members of the public/Incidentally exposed workers and occupationally exposed workers - (Heglig Field)

V. Conclusion

The highest activity concentration and so absorbed dose and annual effective dose was found in block 4 (this is due to existence of scale only in block 4). The highest concentration was found in scale and sand samples extracted from heaters, vessels and tanks during routine maintenance in CPF and different FPFs. It obvious that all samples annual effective dose are less than 20 mSv/y (which is the limit for occupational workers). But four out of five scale samples and five out of six sand samples from Neem field exceed 1mSv/y (which is the limit for public and incidentally exposed workers (locations where those samples were collected are not accessible to public). All sludge samples had AED less than 1mSv/y. All other samples from Heglig, Diffra and Bamboo fields have AED less than 1 mSv/y except sand from heglig field which had AED equal to 2.9 mSv/y.

The highest concentration of radionuclides are found in scale sample, and this may be due to that the main types of scale encountered in oil & gas facilities are sulphate scale such as $BaSO_4$, $SrSO_4$ and carbonate scale such as $CaCO_3$. Radium is chemically similar to barium (Ba), strontium (Sr) and calcium (Ca), hence radium co-precipitates with Sr, Ba or Ca scale forming radium sulphate, radium carbonate and – in some cases – radium silicate.

Limitation and future work

One should consider the limited number of samples collected, and samples randomly collection. So fields survey before sampling can carry out for future work. Also samples radionuclides can be identify using HPGe Detector which had best resolution than NaI (TL).

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Appendix
Some sample sources

A bulk of scale extracted during Routine maintenance

An abandoned pipeline due to heavy scale formation

Sludge inside heater treater

Sand extracted from vessel and put in drums in special closed area