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NATURALLY OCCURRING RADIOACTIVE MATERIALS IN ALGERIA

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ABSTRACT
The NORM characterisation from Algerian oil facilities has been determined by external radiation measurements at the surface of production treatment plants and gamma spectrometry analysis of liquid and solid samples. Background NORM levels were found to range from 0.07 to 0.8 μSv/h. Oil installations and associated equipment NORM levels ranged from 0.1 to 100 μSv/h. Analytical results indicate that ^{226}Ra, ^{214}Bi and ^{214}Pb from ^{238}U series are present in scales and formation water samples. Specific activity ranged from traces to 1000 Bq/g. Uncertainties of both characteristics were typically less than 20 %. According to an extensive world wide literature survey on NORM in oil industry, these results are relatively not negligible.

INTRODUCTION
When oil is extracted from the ground, it is accompanied by solids and formation water. Under certain circumstances, naturally radioactive salts, which are dissolved in the formation waters will precipitate and be incorporated in dense solids that accumulate, either in large volume or, as thin scales that deposit on the internal surfaces of equipments. Deposited sand and sludges may also contain enhanced concentrations of the radioactive substances. Radioactive gas (radon) is also unavoidably extracted and when its decays, the solid "daughters" or "progeny" that result are also incorporated in the solids. The oil industry refers to the solids as NORM (Naturally Occurring Radioactive Materials) and LSA (Low Specific Activity) scales. The formation of the radioactive solids depends on local geological conditions and production methods and is not common to all oilfields. The activity concentration is spatially and temporally variable and may be quite different even from wells producing from the same field at the same time [1]. Typically the solids contain activities that are more than one hundred times greater than the activity concentration found in common rock samples [2]. Nevertheless, the activity concentration is still low by comparison with the specific activity of man-made radioactive sources. Where NORM is present, it may occur downhole, in topside plant production equipment, transport (pipeline) systems, storage facilities, and oil handling sites [3].
In Algeria, there are a number of industrial activities involving naturally occurring radioactive materials. However, the activity concerned is the oil industry. The objective of this study was to identify the hazards produced using radiation survey and sampling techniques.

RADIOLOGICAL CHARACTERIZATION OF NORM
The major oilfields in Algeria include treatment and production installations. Oilfields concerned by the control of NORM hazards were A, B, C and D located in Southern Algeria. The operational radiological monitoring carried out, was gamma radiation level and concentration activity/level.

Gamma radiation level
Gamma radiation level is performed when NORM scales are contained within equipments or tubulars and cannot be sampled. To achieve this, external measurement were taken at the surface (1cm) of flow lines, tubings and equipments. All NORM levels were measured in μSv/h. Automess S/N 90803 instruments with appropriate probes (6150 AD-18 for gamma, 6150 AD-19 for beta and 6150 AD-17 for alpha/Beta/gamma) were used. Prior to each set of measurements, a background reading was undertaken at a position not expected to be influenced by NORM contamination. Approximately 800 NORM measurements were obtained at 100 wells (including water, abandoned and non-producing), 20 manifolds, 40 Separators and 8 Process plants locations and equipments. The uncertainties of the measurements carried out by Automess S/N 90803 instruments were typically less than 20 %. Results obtained are given in Table 1.

Internal measurements of NORM contamination were not performed during this survey since line breaks producing access were not performed during the time frame of this survey.

b- Concentration activity/level

The crude oil, production waters and scales samples were collected from each oilfield under study. The sampling locations were respectively productor wells for liquids, and valves for scale samples. In the laboratory, the samples were conditioned and stored in vials for scales and in sealed polyethylene containers for liquids at least 3 weeks to allow for equilibrium of ^{226}Ra with its decay products. The activity concentrations of natural radionuclides in the samples were determined by low background gamma spectrometry using GeHp detector (10 cm Pb shielding) with a resolution of 2 Kev and relative efficiency of 20 % at 1.33 Mev. The ^{226}Ra concentrations have been derived from the weighted mean of the activities of two photopeaks of ^{214}Pb (295.2, 351.9 Kev) and three photopeaks of ^{214}Bi (609.3, 1120.3, 1764.5 Kev). For ^{232}Th, three photopeaks of ^{228}Ac (338.4, ,911.1, 960.1 Kev) and the photopeaks of ^{212}Pb (286.6 Kev), ^{208}Tl (583.2 Kev) and ^{208}Bi (727.2 Kev) were used in the same way. The concentration of ^{40}K was obtained from the photopeack at 1460.8 Kev.

The energy and efficiency calibrations were carried out using the IAEA reference material and the multigamma solution from the Centre of Radiation Protection and Safety (CRS) with the same geometry as that of the samples. The measurement time ranged from 14 to 24 h. Gamma spectra were analysed by using the GENIPC (1995) software provided by CAMBERA and the software elaborated by the CRS. The uncertainties of the measurements carried out by gamma spectrometry were typically less than 20 %. Important results are given in Tables 2,3 and 4.

RESULTS AND DISCUSSION

Gamma radiation level

The contents of the naturally occurring radionuclides in oil industry in different regions have not yet systematically studied. In the results presented on the external measurements (Table 1), approximately (800) measurements were obtained at 100 wells, 20 Manifolds, 40 separators and 8 process plant locations and associated equipment. Background levels ranged from 0.07 to 0.8 μSv/h. The highests background levels were 0.6 μSv/h and 0.8 μSv/h respectively in A and D oilfields. NORM levels ranged from 0.06 at several wells to 80 μSv/h at D oilfield wells. Levels for the manifolds ranged from 0.5 to 100 μSv/h with the highest NORM levels at the D oilfield north manifold. Process plant and associated equipment levels ranged from 0.1 to 17 μSv/h. Levels for the separators ranged from 0.2 to 21 μSv/h. The NORM levels at storage tanks ranged from 0.07 to 0.20 μSv/h.

The highest NORM levels for this survey were found at the A oilfield. According to the literature data [4], it appears from these results that radiation levels are very significant, especially at A and D production facilities. This is due, on one hand to the difference in the geological formation from which the oil is extracted and on the other hand the chemical incompatibility between the produced water containing Ba^{++}, Sr^{++} and Ca^{++} and injected water containing SO_4^{--}, used to increase the production pressure.

Table 1: **Observed external radiation levels at outside production facilities**

Oilfield	Location measurements with radiation levels (μSv/h)					
	Wells	Manifolds	Plants (Gas)	Separators	Storage Tanks	Scale from valves
A	0.1 – 50	0.1 – 100	0.3 – 0.9	0.2 – 21	0.1 – 0.6	10
B	0.1 – 100	0.5 – 14	0.1 – 4	0.5 – 14	–	12.5
C	0.4 – 1	0.4 – 20	0.4 – 2	0.2 – 3	0.2 – 7	10
D	0.1 – 80	0.12 – 17	0.12 – 0.25	0.1 – 17	0.12 – 0.18	17

Concentration activity/level

The naturally occurring radionuclides identified in Algerian oil industry are given in Tables 2 to 4. From these radionuclides analysis carried out on materials collected from four (04) different oilfields, it has become clear that the radionuclides found in downhole and surface structure installations do not include the primordial nuclides ^{238}U and ^{232}Th. These elements are not apparently mobilised from the reservoir rock, which contains the oil and formation water. The latter contains group II cations Ca, Ba, Sr as well as Ra dissolved from reservoir rock [5,6]. These nuclides have been co-precipitated with group II cations as sulphate and scales onto the inner walls of production and treatment installation equipments. Table 2 shows that the extent of the mobilisation of radionuclides from the different reservoirs and their amounts in produced water varies very strongly between installations and between individual wells. In general more heavy scaling is encountered in oil producing installations with more saline produced water. However, at the D oilfield, the chemical incompatibility, which provides sulphate, scales, does not exist, but the activities are greater than those obtained at A with chemical incompatibility between produced and injected waters. This is due to the presence of carbonated injected water and Ra and its daughters in formation water. For the purpose of the present study it may be taken that the concentrations of NORM in scales cover a range of a few tenths Bq/g up to more than 1000 Bq/g (Table 3). Generally, the activity concentrations of ^{226}Ra in water and oil are lower than in scales. The same applies to ^{214}Pb and ^{214}Bi for all samples. However, it should be noted that although thorium isotopes are not mobilised from the reservoirs. Produced water contains NORM but oil is a virtually free (Tables 2 and 4).

Table 2: **Activity concentration of radionuclides in formation water**

Radionuclides ($Bq.g^{-1}$)	Oil fields			
	A	B	C	D
^{226}Ra	0.51	-	0.02	0.50
^{214}Pb	0.14	-	0.04	0.20
^{214}Bi	0.14	-	0.02	0.30

Table 3: **Activity concentration of radionuclides in scales in different Oilfields**

Radionuclides ($Bq.g^{-1}$)	Oil fields			
	A	B	C	D
^{226}Ra	289.31	-	18.14	645.50
^{214}Pb	289.11	-	10.85	629.05
^{214}Bi	292.50	-	6.50	618.52

Table 4: **Activity concentration of radionuclides in oil in different Oilfields**

Radionuclides ($Bq.g^{-1}$)	Oil fields			
	A	B	C	D
^{226}Ra	0.067	-	0.02	0.05
^{214}Pb	0.066	-	0.02	0.06
^{214}Bi	0.069	-	0.02	0.06

(-): Analysis in progress

CONCLUSION

NORM is acknowledged as a problem in many parts of the word and in our own region. Some industries still appear to be unaware that may be present in their operations. Other activities such as phosphate fertiliser industry and other practices producing NORM have not been covered in this study.

Our results from NORM characterisation in Algerian oil industry reveal that:

- Survey data indicates that NORM is present at several oilfield locations and proper handling and storage procedures are required to protect workers and the environment.
- Gamma spectrometry analysis of scale samples showed that specific activity was relatively not negligible.
- NORM is originated by formation water.

Furthermore, it is concluded that for Algerian oil industry, the naturally occurring radioactive materials, is somewhat small compared to other artificial radioactive material used for various activities in the country. NORM characterisation obtained will be used for an assessment of potential radiological safety and environmental hazards.

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