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Original Article

Assessment of occupational radiation exposure of NORM scales residues from oil and gas production

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ABSTRACT

Radiological hazards from external exposure of naturally occurring radioactive materials (NORM) scales residues, generated during the extraction process of oil and gas production in southern Algeria, are evaluated. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were measured using high-purity gamma-ray spectrometry (GeHP). Mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, found in scale samples are 4082 ± 41, 1060 ± 38 and 568 ± 36 Bq kg^{-1}, respectively. Radiological hazard parameters, such as radium equivalent (Ra_{eq}), external and internal hazard indices (H_{ex}, H_{in}), and gamma index (I_{γ}) are also evaluated. All hazard parameter values were greater than the permissible and recommended limits and the average annual effective dose value exceeded the dose constraint (0.3 mSv y^{-1}). However, for occasionally exposed workers, the dose rate of 0.65 ± 0.02 mSv y^{-1} is lower than recommended limit of 1 mSv y^{-1} for public.

1. Introduction

Natural radioactivity from uranium and thorium always exists in the human environment. They are found in soil, rocks, water, and food, and can be observed in the human body [1].The main radionuclides are uranium (^{238}U), fissionable uranium (^{235}U) and their descendants, the family of thorium- 232 (^{232}Th) and potasium-40 (^{40}K) [2]. This radioactivity is not hazardous, as long as it remains in its natural state [3]. However, certain work activities, such as mining and oil or gas production, which involve different physicochemical processes, can significantly increase exposure to this radiation and need to be regulated. This increase in exposure is due to naturally occurring radioactive materials (NORM) [4]. NORM generally consists of natural elements and their daughters, such as radium and radon isotopes. Those materials can cause health hazard to the population and pollute the environment. Danger and pollution levels depend on the distribution and exposure of materials, as well as chemical or physical processes [5]. It can be detected and measured through ionizing radiation using nuclear techniques, but it cannot be detected by the human body [6].

Since the 1930s, NORM have been recognised as being present in oil and gas production, petroleum reservoirs and processing facilities [7].

Each year, the production of NORM residues in the oil and gas industry is estimated to be 150,000 cubic meters. This amount depends on several, factors such as geological formation, type of production, age of the industry, and the sampling site [8]. NORM accumulates at different locations in the oil and gas industries. Among these locations are the wellheads, pumps and separator vessels. The build-up of NORM may take the form of scales, sludge, scraping and other wastes [9]. NORM scale formations are due to the precipitation of alkaline earth metal sulfates and carbonates caused by the solubility of change. Variations in sulfate and carbonate solubilities are associated with temperature and pressure variations, gas expansion due to pipeline diameter change, varying flow (transition between laminar and turbulent), and injection of incompatible water [10]. Water injected into reservoirs to maintain pressure is the main cause of scale formation [11]. Generally, scales found in oil and gas production are sulfate scales, such as barium sulfate (BaSO4), and carbonate scales, such as calcium carbonate (CaCO3). Radium (Ra), with the same chemical proprieties as

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barium (Ba) and calcium (Ca), co-precipitates with them, forming trace impurities fixed within the parois [12]. Scale residues can contain extremely high quantities of radioactive materials, unlike soils and rocks [13]. For example, in the oil and gas production, the activity concentration of ^{226}Ra from scale NORM residues ranges from 185 to several tens of thousands of Bq kg^{-1} [8],whereas that of rocks and soil is approximately 18.5–185 Bq kg^{-1} [14].

Personal irradiation in storage areas and contamination during scales removal procedures are the main hazards associated with scales [15]. A high concentration of NORM on equipment walls leads to external exposure, and if physical protection measures are not taken, dust inhalation can occur [5].

In the oil and gas industry, workers are typically exposed to gamma radiation, dust inhalation, and skin beta contamination [16], but workers respect physical protection measures, such as wearing masks during cleaning and equipment maintenance. Therefore, dust inhalation can be minimized and control is necessary to limit exposure [2]. The emission of gamma radiation is linked to the concentration of NORM; however, the correlation between these two is very difficult because some gamma rays emitted do not pass through the walls of the equipment; moreover, the distribution of NORM in the equipment can vary [17]. This can be mitigated by applying chemical descaling methods, in which the solubility of materials can be achieved using acid solutions [13]. The guidelines developed for these NORM will also allow better control of the NORM to protect people and the environment [12].

2. Materials and methods

2.1. Sample collection and preparation

NORM residue scale samples were collected from the oil and gas industry during reviews and cleaning operations in different facilities (e.g., separator tanks, water treatment vessels, gas treatment, pipelines …). They accumulate at the bottom of the facility. For each residue from one facility, five samples were collected and thoroughly mixed to obtain homogenous and representative quantities.

In the laboratory, the samples were dried in an oven at 105 °C for at least 24 h until a constant weight was achieved. They were then sieved through a 200 μm diameter sieve. Therefore, their particle sizes were less than or equal to 200 μm. To allow for the equilibrium of ^{226}Ra with its decay products, such as ^{214}Bi and ^{214}Pb, the collected samples were stored for at least one month in 250 cc polyethylene bottles with the same dimensions as the standard sample. Samples were weighed and manufactured using S1, S2, S3, S4, and S5.

2.2. Gamma-ray spectrometry

Gamma ray measurements were carried out using a high-purity germanium (HPGe) detector, model GC-3018 (10 cm Pb shielding), manufactured by Canberra, used for routine gamma spectrometry in the radioisotope laboratory of CRNA (Nuclear Research Center of Alger) with a resolution of 1.8 keV at 1332 keV for ^{60}Co and 0.875 keV at 122 keV for ^{57}Co, a relative efficiency of 34.7%, and an active volume of 250 cm^3. Efficiency calibration was performed with standard ^{152}Eu, ^{133}Ba, ^{241}Am sources mixed with the soil matrix in a plastic bottle. The genie 2000 software was used to acquire and treat the collected data.

All concentration activity values are obtained by equation (1) [18].

$$A = \frac{N - N_B}{I_\gamma . \varepsilon(E_\gamma) . t . m} \tag{1}$$

N and N_B are the integral; and background surface, respectively (counts); , I_γ and $\varepsilon(E\gamma)$ are the gamma ray intensity and the detection efficiency of the radionuclide (%), respectively; t is the time acquisition (s); and m is the sample mass (kg).

The minimum detectable activity concentration (A_{MDC}) was calculated using follow equation (2) [19].

$$A_{MDC}(Bq/kg) = \frac{F_C . \sigma_{NB}}{\varepsilon(E_\gamma) . I_\gamma . t . m} \tag{2}$$

Where F_C is the statistical coverage factor equal to 1.64 and σ_{NB} is the standard deviation of the background in the region of interest and is the square root of the number of counts for the background spectrum.

The values of 0.6, 0.3, and 6.2 Bq kg^{-1} were obtained as the minimum detectable activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The mean value of A_{MDC} for all interested radionuclides was estimated at 1.4 Bq kg^{-1}.

2.3. Doses assessment and radiological hazards parameters determination

To estimate the exposure risk of gamma radiations from NORM scale residues, received doses such as the absorbed dose rate (D) and annual effective dose equivalent (AED) were estimated.

2.3.1. Doses assessment

2.3.1.1. Absorbed dose rate (D_{abs}). The absorbed dose rate in the air measured at 1 m above ground level from the ground surface which assesses the external exposure of gamma radiation of ^{266}Ra, ^{232}Th and ^{40}K can be determined using equation (3) [20].

$$D_{abs}\left(nGy.h^{-1}\right) = 0.462A_{Ra-226} + 0.621A_{Th-232} + 0.0417A_{K-40} \tag{3}$$

A_{Ra}, A_{Th} and A_K represent the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The absorbed dose limit was 0.059 mGy h^{-1} [20].

2.3.1.2. Annual effective dose equivalent (AED). The annual effective dose equivalent of γ-rays for adult exposure was estimated using equation (3) [13,15]. This equation (4) takes into account the conversion factor F_γ of 0.7 Sv Gy^{-1} used to toggle of dose rate to annual effective dose and the annual exposure time (T_{exp}) which is equal 1820 h (7h × 5d × 52w) [21].

$$AED = D_{abs} \times F_\gamma \times T_{\exp} \tag{4}$$

For occupational exposure, the AED (mSv y^{-1}) resulting from the absorbed dose values (D) can be calculated using equation (5):

$$AED\left(mSv.y^{-1}\right) = D_{abs}\left(nGy.h^{-1}\right) \times 0.2 \times 1820\left(h.y^{-1}\right) \times 0.7\left(Sv.Gy^{-1}\right) \times 10^{-6} \tag{5}$$

where 0.2 represent the occupational factor. The annual effective dose of γ-rays must be less than the global average of 0.460 mSv y^{-1} [20].

2.3.1.3. Annual gonadal dose equivalent (AGDE). The annual equivalent dose received each year by gonads for the exposed population is represented by AGDE [22]. This dose, estimated with the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the NORM residues scales was calculated using equation (6) [23].

$$AGDE\left(\mu Sv.y^{-1}\right) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \quad (6)$$

2.3.2. Radiological hazards parameters

As 98.5% of the radiological effects linked to ^{238}U decay series are due to ^{226}Ra, the contribution from ^{238}U is replaced by that of ^{226} Ra. Usually, in petroleum materials, the natural activity concentration is determined by ^{226}Ra, ^{232}Th, and ^{40}K [21]. To assess radiological hazards from γ-rays due to NORM residues scales, parameters such as radium-equivalent (Ra_{eq}), external and internal hazards indices (H_{in}, H_{ex}), gamma index (Iγ) and activity utilization index (AUI) were calculated. These indices contribute significantly to health risks if they are greater than the world average [24].

2.3.2.1. Radium equivalent (Ra_{eq}). To account for the non-uniform distribution of radionuclides in the NORM samples, radioactivity was defined in terms of radium-equivalent which is one of the most calculated parameters to determine radiological hazards [25]. This is defined by Equation (7).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (7)$$

In this formula, A_{Ra}, A_{Th} and A_K represent activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively and the base estimation is 370 Bq kg^{-1} for ^{226}Ra, 259 Bq kg^{-1} for ^{232}Th, and 4810 Bq kg^{-1} of ^{40}K to provide the same gamma radiation dose equivalent. The limit recommended for the radium-equivalent is 370 Bq kg^{-1} [26].

2.3.2.2. External and internal hazards indices (H_{ex}, H_{in}). The external hazard index is used to evaluate the external hazard due to emitted γ-rays. It is produced from the Ra_{eq} expression and is defined by Equation (8) [26].

$$H_{ex} = \frac{A_{Ra-226}}{370Bq.Kg^{-1}} + \frac{A_{Th-232}}{259Bq.Kg^{-1}} + \frac{A_{K-40}}{4810Bq.Kg^{-1}} \leq 1 \quad (8)$$

The internal hazard index was used to assess internal exposure to carcinogenic radon. It was estimated using Equation (9) [22].

$$H_{in} = \frac{A_{Ra-226}}{185Bq.Kg^{-1}} + \frac{A_{Th-232}}{259Bq.Kg^{-1}} + \frac{A_{K-40}}{4810Bq.Kg^{-1}} \leq 1 \quad (9)$$

These two values must be less than 1 to avoid being considered.

2.3.2.3. Gamma index Iγ. Index Iγ permits the examination of the concentrations of natural radionuclides to assess the risks due of gamma radiation in the NORM sample. It is a good test parameter for radionuclides that may have harmful health effects. The gamma index was determined using Equation (10) given by the European Commission [27].

$$I_\gamma = \frac{A_{Ra-226}}{300Bq.kg^{-1}} + \frac{A_{Th-232}}{200Bq.kg^{-1}} + \frac{A_{K-40}}{3000Bq.kg^{-1}} \quad (10)$$

A_{Ra}, A_{Th} and A_K represent the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

European commission suggests that the exemption dose criterion of 0.3 mSv y^{-1} correspond to Iγ lower than 0.5 and the 1 mSv y^{-1} exemption dose correspond to Iγ lower than 1. In other words, these dose criteria are accepted if Iγ is less than unity corresponding to the effective annual dose [27].

Hence, it is recommended to controls dose between 0.3 and 1 mSv [28].

2.3.2.4. Activity utilization index (AUI). The activity utilization index (AUI) represents the ambient air dose rate of several combinations of the three radionuclides (^{40}K, ^{232}Th, and ^{226}Ra) in the scale NORM sample. It is calculated from Equation (11) which uses suitable conversion factors for each corresponding radionuclides [23].

$$AUI = \frac{A_{Ra-226}}{50Bq.Kg^{-1}}Cf_{Ra} + \frac{A_{Th-232}}{50Bq.Kg^{-1}}Cf_{Th} + \frac{A_{K-40}}{500Bq.Kg^{-1}}Cf_K \quad (11)$$

Cf_{Ra} (0.462) Cf_{Th} (0.604) and Cf_K (0.041) are the appropriate conversion factors from the measured activities of the radionuclides to the dose rate of gamma radiation in the air [27]. In the NORM residue scale, the permissible levels for specific activities per unit weight of ^{226}Ra, ^{232}Th and ^{40}K are 50, 50 and 500 Bq kg^{-1} respectively [24,27].

3. Results and discussion

3.1. Activity concentration

The activity concentration of each sample (S1 to S5) was calculated according to Equation (1) (Table 1). Gamma lines of 295.2 and 351.9 keV for ^{214}Pb and 609.3 keV of ^{214}Bi are used to determine of ^{226}Ra activity concentrations. Gamma lines of 238.6 keV for ^{212}Pb, 911.6 keV for ^{228}Ac and 583.18 keV of ^{208}Tl, respectively, were used to determine ^{232}Th activity concentration. Finally, ^{40}K activity concentration was determined using a single gamma line (1460.7 keV).

Fig. 1 reports graph of ^{226}Ra, ^{232}Th and ^{40}K activity concentration values radionuclides in the samples scale NORM.

^{226}Ra activity concentration values varied from 25 to 15798 Bq kg^{-1} with a mean of 4082 Bq kg^{-1}. These values are in agreement with the literature which shows values of up to 15000 Bq kg^{-1} [29,30].

For ^{232}Th, the activity concentration values were in the range of 15–4010 Bq kg^{-1} with a mean value of 1060 Bq kg^{-1}, which is lower than that of ^{226}Ra. In contrast, ^{40}K activity concentration values (27–1697 Bq kg^{-1}) were much lower than those for ^{226}Ra and ^{232}Th, with a mean value of 568 Bq kg^{-1}.

^{226}Ra activity concentrations obtained in samples 3 and 4 (15798 and 4137 Bq kg^{-1} respectively) were higher than those of the other samples because they were collected from the separator and water treatment vessels. This high concentration difference of NORM between the samples can be explained by two factors; the accumulation of NORM scales in the separator and the chemical properties of the radium.

NORM scales residues accumulate in the separator tank, which constitutes the first phase of the oil and gas treatment and chemical property of radium to dissolve in water means that the concentration level of radium remains very high in the water treatment vessels. The radium in these facilities can reach 1000 kBq kg^{-1} [13].

As previously reported, ^{226}Ra is the major radionuclide in NORM

Table 1
Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in scale NORM residues.

Sample	Activity concentration (Bq Kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K
S1	235 ± 11	60 ± 3	493 ± 24
S2	215 ± 48	45 ± 8	1697 ± 122
S3	15798 ± 96	4010 ± 87	326 ± 12
S4	4137 ± 49	1172 ± 86	298 ± 12
S5	25 ± 1	15 ± 8	27 ± 12
Range	25–15798	15–4010	27–1697
Mean	4082 ± 41	1060 ± 38	568 ± 36

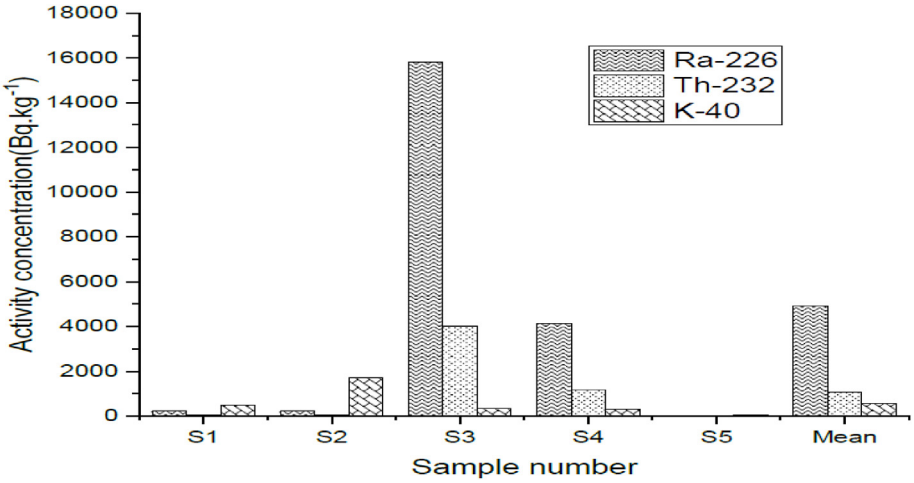

Fig. 1. Activity concentration values of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the samples.

residues in the oil and gas industries. Its enhanced ^{226}Ra activity over ^{232}Th and ^{40}K, is due to its physicochemical properties; such as longer half-life and lower solubility [31]. The mean value for ^{226}Ra (4082 Bq kg^{-1}) was higher than the world average radioactivity (300 Bq kg^{-1}) [32]. For ^{40}K, the activity concentration values did not exceed the fixed rejection limit of 17000 Bq kg^{-1}, which is the natural abundance of potassium [20].

Therefore, these residues are a significant source of waste. A comparison between the results of this study and those other studies is presented in Table 2.

3.2. Evaluation of some NORM scales radiation hazards indices

3.2.1 Radium equivalent, calculated from equation (7) and reported in Table 3 is ranged between 359 and 21557 Bq kg^{-1} with a mean of 5642 Bq kg^{-1}. The mean value was greater than the recommended maximum value of 370 Bq kg^{-1} [5,25]. Consequently, these residues can cause radiological hazards in occasionally exposed workers. Figure (2) shows these results.

3.2.2 External and internal hazard (H_{ex}, H_{ex}) (eqs. 8 and 9) index, respectively. Table 3 shows that these indices range between 0.96 — 58.2 and 1.33–101 respectively. The H_{ex} and H_{in} values exceed the unity for all simple except for the sample 1. The average values are 17.4 ± 0.45 and 30.6 ± 0.62 respectively. These values also show that the NORM scales produced represent a significant radiological hazard for workers [35,36].

3.2.3 Gamma hazard index, calculated from Equation (10), ranged from 1.24 to 72.8 with an average of 19.1 This value, which is linked to the annual effective dose of the public (1 mSv y^{-1}), exceeds unity.

Table 3
NORM scales radiation hazards indices values.

Samples	Ra_{eq} (Bq kg^{-1})	H_{ex}	H_{in}	I_γ	AUI
S1	359 ± 18	0.96 ± 0.05	1.50 ± 0.08	1.24 ± 0.06	2.93 ± 0.14
S2	410 ± 231	1.10 ± 0.62	1.33 ± 0.88	1.50 ± 0.80	2.66 ± 1.96
S3	21557 ± 230	58.2 ± 0.62	101 ± 0.88	72.8 ± 0.79	194 ± 1.95
S4	5836 ± 230	15.7 ± 0.62	26.8 ± 0.88	19.7 ± 0.79	52.4 ± 1.95
S5	48 ± 14	11.2 ± 0.36	22.4 ± 0.36	13.8 ± 0.47	38.4 ± 1.07
Mean	5642 ± 145	17.4 ± 0.45	30.6 ± 0.62	19.1 ± 0.58	50.6 ± 1.42
Range	48–21557	0.96–58.2	1.33–101	1.24–72.8	2.66–194
Limit	370 Bq kg^{-1}	≤1	≤1	≤1	= 1

3.3. Evaluation of some doses

3.3.1 Results of dose rates calculated from equation (3) are displayed in Table 4 and illustrated in figure (3), and ranged between 166 and 9803 nGy h^{-1}, with an average of 2568 nGy h^{-1}.These values are greater than the limit prescribed by the United Nations Scientific Committee on the Effects of UNSCEAR atomic radiation (57 nGy h^{-1}) for workers in the regions containing gamma radiation [38]. The average value is due to the higher activity concentration of ^{226}Ra in Sample S3. So, the later was a major contribution to the dose rate.

3.3.2 Annual effective dose of workers comes from external exposure to gamma radiation and dust inhalation in the absence of

Table 2
Activity concentration of^{226}Ra and^{232}Th in Algeria and others countries.

Country	Average activity concentration (Bq kg^{-1})		References
	Ra-226	Th-232	
Egypt	(mean 11700)	(mean 4200)	[13]
Brazil	77,900–2110,000 (mean 897,500)	101,500–1550,000 (mean 679,500)	[33]
Egypt	14,100–28,900	5000–9500	[34]
Egypt	493,000–519,000	1080–2090	[31]
Egypt	1684–2197	574–696	[27]
Syria	300–1,520,000 (mean 174,000)	600–868,000 (mean 91,000)	[27]
Algeria	1000–950,000	–	[5]
Turkey	6–10	< MDA	[35]
KSA (Riyadh city refinery tanks)	0.8 —– 1.5	0.1–3.1	[36]
Ghana	28–48	17–40	[37]
Algeria	25.42–15798.40 (mean 4904.79)	44.63–4010.00 (mean 1060.13)	This study

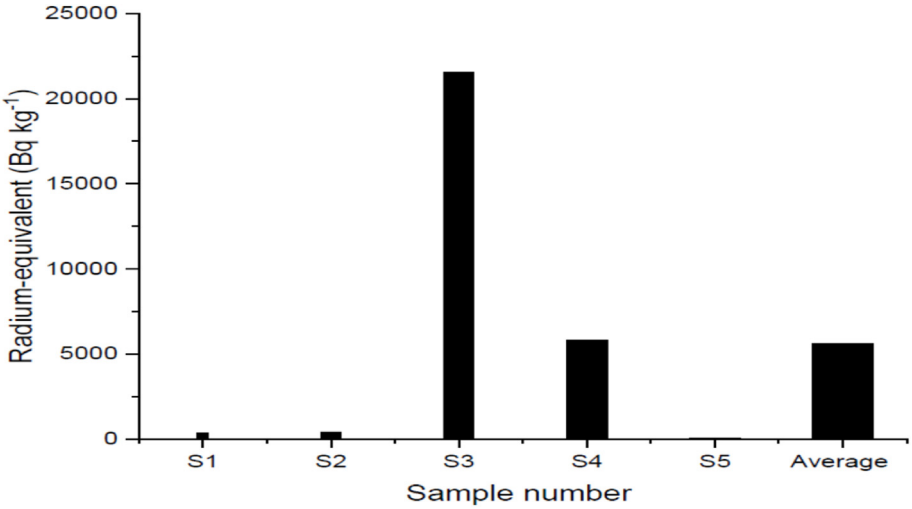

Fig. 2. Radium equivalents for scale NORM residues samples.

Table 4
NORM scale residues dose values.

Samples	D_{abs} (nGy h^{-1})	AED (mSv y^{-1})	AGDE (μSv y^{-1})
S1	166 ± 83	0.04 ± 0.01	1132 ± 56
S2	198 ± 10	0.05 ± 0.02	1385 ± 70
S3	9803 ± 104	2.49 ± 0.03	65680 ± 699
S4	2652 ± 104	0.67 ± 0.02	17776 ± 698
S5	22 ± 6	0.01 ± 0.01	148 ± 40
Mean	2568 ± 61	0.65 ± 0.02	17224 ± 512
Range	166–9803	0.01–2.49	148–65680
Limit	57 nGy h^{-1}	≤1 mSv y^{-1}	≤ 300 μSv y^{-1}

physical protection measures and adequate controls during maintenance procedures [13]. In oil and gas production, the hazardous inhalation of radioactive dust is lower for workers. Therefore, we evaluated only the annual effective dose from to external gamma radiation exposure. These values, calculated from Equation (5), are presented in Table 4. It is ranged from 0.01 to 2.49 with an average of 0.65 mSv h^{-1} exceeding dose constraint (0.3 mSv) but does not exceed the limit recommended for the public (1 mSv y^{-1}) [39]. In sample S3, the annual effective dose is higher than the safety limit (1 mSv y^{-1}) recommended by UNSCEAR, but does not exceed the tolerable limit for workers occasionally exposed which is 5 mSv y^{-1} [40]. Knowing that protection measures, such as distance and attenuation, cannot be applied under these conditions, these results show that it is necessary to limit the exposure time, either by reducing the time residence of workers or performing the dose-sharing technique. The goal is to maintain the exposure of workers as low as possible (ALARA).

3.3.3 Annual gonadal dose equivalent calculated from equation (6) and presented in Table 4 range of 148–65680 with an average of 17224 μSv y^{-1}. These values, exceeding the recommended limit (300 μSv y-1), show that there is high exposure of gonads in this oil and gas production.

4. Conclusion

In this study, the activity concentrations of ^{228}Ra, ^{232}Th and ^{40}K and some radiation parameters were calculated to assess the radiological hazards from external exposure of workers in the oil and gas industry in southern Algeria. Gamma spectrometry revealed the presence of natural radioactive ^{238}U and ^{232}Th radionuclide chains and ^{40}K radionuclides.

From the results of the specific activities obtained from these materials, it appears that the ^{226}Ra and ^{232}Th activity concentrations are very high in samples S3 and S4 (15798 ± 96 and 4010 ± 87 Bq kg^{-1}, 4137 ± 49 and 1172 ± 86 Bq kg^{-1} respectively), exceeding the safe limit recommended by the ICRP. This indicated that the

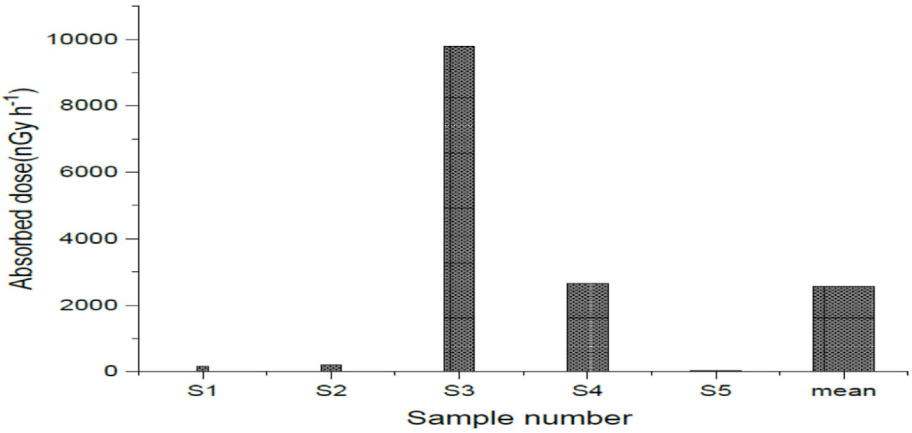

Fig. 3. Absorbed dose rates for scale NORM residues sample.

first production equipment were responsible for the level of radioactivity in oil and gas production. It can be concluded that this residue should be considered radioactive waste and managed accordingly. ^{40}K, on the other hand, is of the order of natural background noise in soils.

All mean hazard parameters are greater than the recommended safety limit, and the estimated dose rates show that long-term accumulation of NORM scale residues may lead to possible radiological hazards in the future.

Therefore, during the cleaning and maintenance process in the oil and gas production, special and save methods should be used, such as chemical treatment of NORM scale residues, store NORM residues in underground bunkers and regulatory control to reduce personal exposure and prevent contamination of the environment.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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