Assessment of Naturally Occurring Radioactive Materials (NORMs) in soils from the Kuçova oilfield, Albania

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Abstract: - Oil and gas exploration activities belong to the industrial sectors involving the production of NORMs because of the enhancement in radium isotopes concentrations recorded in industrial residues. According to the European recommendations of the new Basic Safety Standards for radioprotection, each member state shall identify NORM industries and estimate their environmental and human impact. In this framework, the concentrations of uranium and thorium decay series, as well as of ^{40}K were measured in soil, sludge and oil sand samples collected in the Kuçova oilfield (Albania) by using a high resolution gamma-ray spectrometer (HPGe). The levels of radioactivity in soil samples were found to be comparable with the world mean activity concentrations. Oil sand samples showed the highest ^{40}K and ^{226}Ra activity concentrations, indicating the presence of shale traps, which are generally rich in natural radionuclides. No strong evidences on disequilibrium were observed from the ratio of ^{228}Th and ^{228}Ra activity concentrations. The concentration of

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radionuclides in produced water and crude oil were found to be minimum detectable activities. From the radiological point of view, the excess of annual effective dose rate from industrial residues was estimated to be much lower than the recommended limits for population and workers.

Key-Words: -NORM; Natural radioactivity; Oil and gas extraction; Gamma-ray spectrometry, Radium isotopes; Kuçova oilfield; Albania

1 Introduction

Naturally Occurring Radioactive Materials (NORMs) are residues enriched with radionuclides of natural origin as a consequence of industrial processes [1]. The main sources of natural radioactivity are ^{238}U, ^{232}Th and their progenies and ^{40}K radionuclide which are present in the Earth's crust, in soils and in waters at varying abundances. The concentrations in soil and water can be enhanced by industrial processes, such as oil and gas extraction, production of phosphate fertilizers, coal mining and combustion, cement production, which are listed in [2] as industrial sectors involving NORMs. Therefore, the identification and monitoring of industrial processes involving NORMs should be carried out by countries for assessing the exposure of workers or members of the public.

During the last years, particular interest has been devoted to unconventional methods of shale gas extraction known as "fracking", which causes NORMs to be brought to the surface as part of the flowback and production brine. In oil and gas industry, specific attention has been dedicated to the contamination of oil equipment and oilfield environment with NORMs (e.g. scale, sludge and produced water). In particular, these residues are enriched in radium isotopes originating from uranium and thorium present in reservoir rocks. Indeed, while both uranium and thorium present in hydrocarbon reservoir rocks are essentially insoluble under reducing conditions, their progenies $^{226/228}$Ra concentrate in formation waters. For this reason, $^{226/228}$Ra are unsupported by the long-lived uranium and thorium parent radionuclides and, due to their half-lives of 1600 yr and 5.75 yr respectively, they tend to accumulate in formation water. The produced water (i.e. water brought in surface) may contain in solution various cations like barium, calcium, and strontium, as well as sulfate and carbonate anions, whose concentrations depend on the solubility of the original salts, which in turn is influenced by cation/anion ratio, pH, temperature and pressure. Since radium is chemically similar to Ba, Ca and Sr, as produced water is brought in surface and temperature tends to drop, it can consequently co-precipitate as radium sulfates or radium carbonates: this can lead to the subsequent formation of scales, depending on the concentration of ionic species in the produced water [3, 4]. ^{226}Ra and ^{228}Ra concentrations in scale can reach values as high as 1500×10^4 Bq/kg and 280×10^4 Bq/kg respectively, as reported by **IAEA** [5], but typically highly variable concentrations are observed [6].

The petroleum exploration and production in Albania began in early XX century with the construction in the Kuçova area of the oldest and the second biggest oilfield of the country. The primary oil extraction technique in Albania uses beam pumps, which allow for exploiting the pressure of the gas in the reservoir to force oil out and into the well. Since statistically 25 to 95% of the original oil has not been extracted after the well consumed the reservoir's natural drives, water and steam-drive improved recovery generally follows, in order to raise fluid level and pressure in the wellbores and reduce oil viscosity. Recently, several secondary recovery techniques, such as water and steam injection, have been employed in pilot wells in the Kuçova oilfield in order to increase the oil production rate. Several environmental studies have shown that the release of produced water in oil well surroundings, in decantation plants and in oil spillage sites can result in serious soil, water and air contamination due to BTEX compounds or crude oil [7, 8, 9]. However, there is a leak of data on the possible soils contamination and pollution with NORMs associated to the extraction processes of oil and gas in Albania.

This study is the first attempt to investigate the radioactivity concentration in soils of the Kuçova oilfield surroundings and in the oil industry by-products, such as oil-sands and produced water. The Kuçova oilfield is located in the South Central region of Albania, approximately 30 kilometers northeast of the Patos Marinza field. The field was discovered in 1928 and had over 1,700 wells drilled (with a typical maximum depth of approximately 1500 m), resulting in a cumulative production of 23.2 million barrels of heavy oil (13-16° API gravity) by the end of 2006. The actual oil extraction process may produce a weak

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enhancement of natural radioactivity, since moderate temperature variations and low amounts of produced water are involved. Considering that secondary recovery techniques may result in relatively high enhancement of natural radioactivity in residues, these data will provide precious baseline information concerning the presence of natural radioelements in the investigated areas. This will allow also for an estimate of the environmental and human impact of oil industrial activities on the Kuçova area. Moreover, these results will open the way to possible implementations in the Albanian legislative framework of the European recommendation regarding the Basic Safety Standards for protection against the dangers arising from exposure to ionizing radiation [2].

2 Material and methods

2.1 Geological setting of the study area

The onshore petroleum exploration in Albania is mainly located in the Ionian Unit (ION) and in the Durrës basin, also called the Peri-Adriatic Depression (PAD) (Fig. 1). Their hydrocarbon potential is mainly related to the Neogenic molasse deposits located in the peripheral southeastern part of the PAD. These deposits are characterized by the presence of transgressive sequences on the top of underlying Mesozoic and Paleogene limestones. The oil currently explored in the Neogenic reservoirs results from a secondary migration from underlying limestone reservoirs (ION). In fact, the generation in situ of hydrocarbons was precluded in the molasse deposits. As a result of the lithological changes, the oil was accumulated mainly in stratigraphic traps of the PAD clastic deposits, which comprise a considerable number of sandy-clayey mega-sequences. In some cases these megasequences become more complete, beginning with conglomerates and clastic limestones and ending on the top with clays, shale, coal or gypsum. The thickness of the molasses increases from southeast to northwest, reaching a maximum of 5000 m [10].

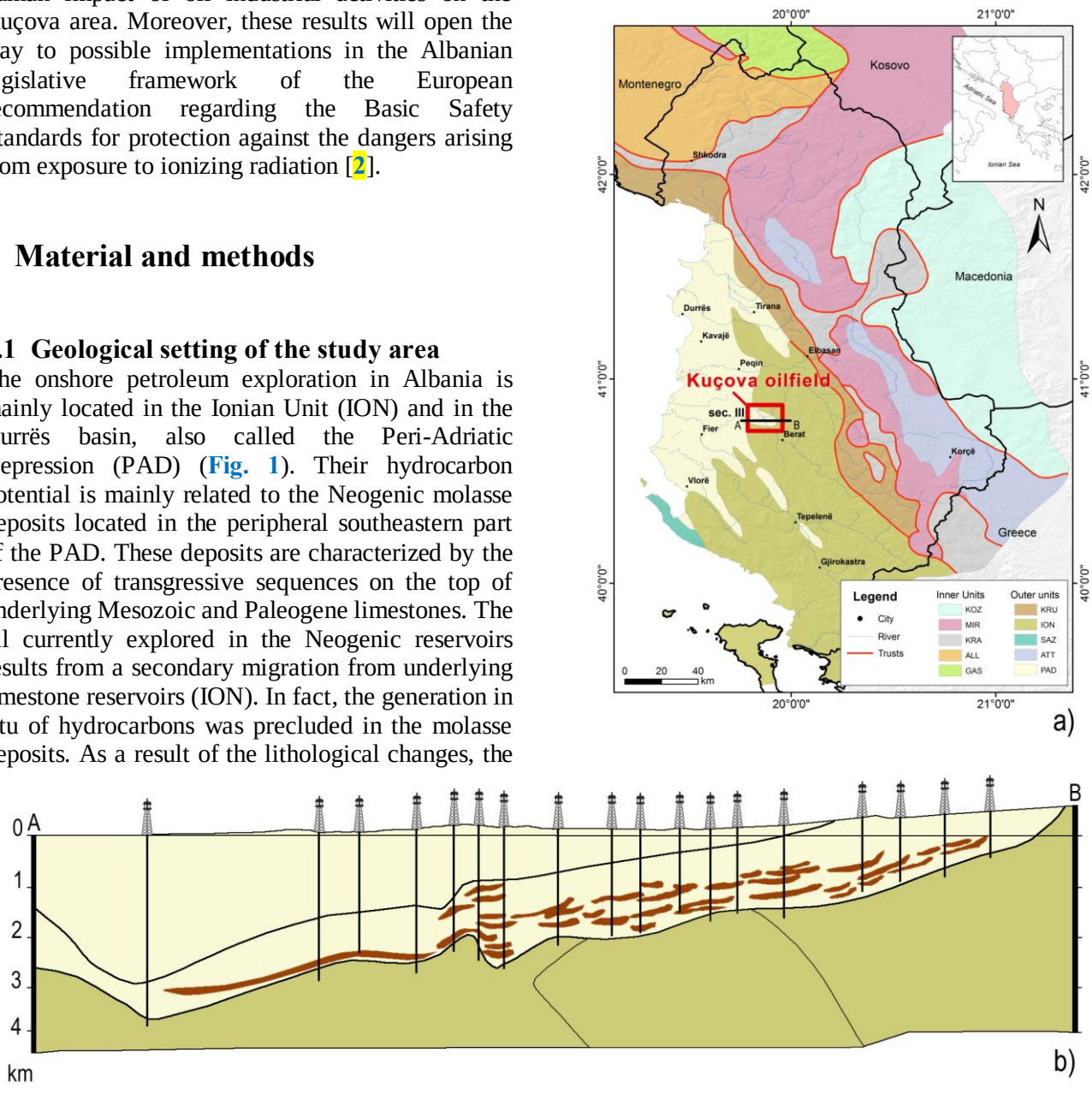

Fig. 1. a) Simplified geological map of Albania, modified from Havancsák et al [11]. Geological units labels are as follows: AAL: Albanian Alps unit; ATT: Albanian-Thessalian trough; GAS: Gashi unit; ION: Ionian unit; KOZ: Korabi unit; KRA: Krasta unit; KRU: Kruja unit; MIR: Mirdita unit; PAD: Periadriatic depression; SAZ: Sazani unit; VER: Vermoshi unit. The red rectangle highlights the location of the Kuçova oilfield. b) The cross section III modified from Silo et al [10] is obtained by a seismic line located in the Kuçova region. PAD and ION are in yellow and green, consistently with panel a).

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2.2 Sample collection and preparation

A total of 21 soil samples were collected at 0-10 cm depth in the surroundings of oil wells in the Kuçova oilfield (Fig. 2). Furthermore, 10 samples of oil sands and 3 samples of sludge (mixture of crude oil, oil sand and soil) were collected during the periodical pipe cleaning process. Samples were homogenized to a grain size of less than 2 mm and dried for at least 24 h at a temperature of 105°C till reaching constant weight. Produced water and crude oil were collected in polyethylene bottles directly from the decantation plants.

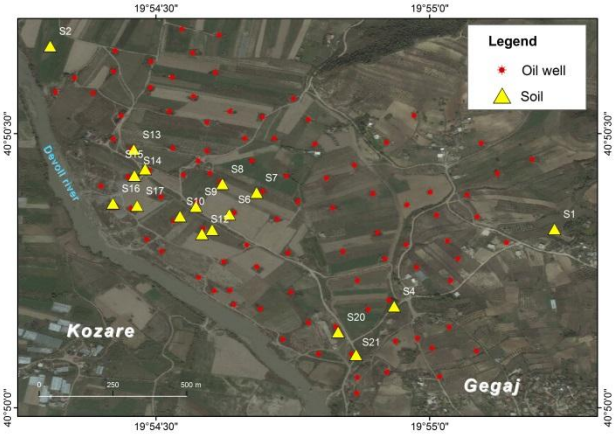

Fig. 2. Approximate locations of soil samples collected in the study area.

Samples were then transferred in cylindrical polycarbonate boxes of 180cm^3 volume and sealed hermetically. Samples were left undisturbed for at least four weeks prior to be measured with the HPGe gamma spectrometer, in order to establish radioactive equilibrium in the ^{226}Ra decay chain segment.

2.3 Gamma-ray spectrometry measurements

Samples were measured at the Laboratory of Nuclear Technologies Applied to the Environment at the University of Ferrara (Italy), using the MCA_Rad system described in Xhixha et al [6]. The fully automated spectrometer consists of two 60 % relative efficiency coaxial p-type HPGe gamma-ray detectors, with an energy resolution of ~1.9 keV at 1332.5 keV (^{60}Co). The absolute full energy peak efficiency of the MCA_Rad is calibrated using certified standard point sources (^{152}Eu and ^{56}Co). The overall uncertainty in the efficiency calibration is estimated to be less than 5%.

The radionuclides studied in this work are ^{226}Ra, ^{228}Ra, ^{228}Th and ^{40}K. The presence of ^{137}Cs in soils was also investigated. The ^{226}Ra activity concentration was determined by analyzing the two main gamma emissions of radon progenies ^{214}Pb (at 352 keV) and ^{214}Bi (at 609 keV) and calculating the weighted average. ^{228}Ra was determined through its direct progeny ^{228}Ac gamma emissions (at 338 keV and 911 keV). The ^{228}Th activity concentration was determined by analyzing the two main gamma emissions of radon progenies ^{212}Pb (at 239 keV) and ^{208}Tl (at 583 keV). The activity concentration of ^{40}K and ^{137}Cs were determined from their respective gamma emissions at 1460 keV and 662 keV. The acquisition time was set to 4 hours for soil, oil sand and sludge samples and 24 hours for produced water and crude oil samples.

2.3.1 Quality control

Certified reference material containing contaminated bulk soil from Syrian oilfield (IAEA-448) [12] was used for quality control. In general ^{226}Ra, ^{228}Ac, ^{212}Pb, ^{208}Tl and ^{40}K activity concentrations agreed within 3-10% with the reference values.

3 Results and discussion

3.1 Activity concentrations

The results on ^{226}Ra and ^{228}Ra activity concentrations with ±1σ standard deviation (in Bq/kg) in soil, oil sand and sludge samples are shown in Table 1. The activity concentrations of ^{226}Ra are generally higher at ±1σ standard deviation for oil sand samples respect to soil samples. On the other hand, the activity concentrations of ^{228}Ra in oil sands are typically comparable within ±1σ standard deviation with concentrations in soil samples. However, regarding the environmental legacy issue, only ^{226}Ra has long-term concern, since after 25 year only approximately 5% of ^{228}Ra still remains.

Several studies on activity concentrations in soil samples from oilfields show a great variability for ^{226}Ra and ^{228}Ra, respectively up to 9 Bq/kg and 11 Bq/kg in Tunisia [13], 42 Bq/kg and 28 Bq/kg in Kuwait [14], 52 Bq/kg and 34 Bq/kg in Nigeria [15], 248 Bq/kg and 29 Bq/kg in Nigeria [16], 10×10^3 Bq/kg and 260 Bq/kg in Canada [17], and 438×10^3 Bq/kg and 987×10^3 Bq/kg in Egypt [18]. The highest values of activity concentration in soil samples were reported in cases of study where higher concentration of ^{226}Ra and ^{228}Ra in scale was present. In cases of study similar to the Kuçova oilfield or when lower concentration of ^{226}Ra and

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^{228}Ra in scale was involved, the concentrations of ^{226}Ra and ^{228}Ra in soil samples are comparable to our measurements.

Table 1. Concentration expressed in Bq/kg at ±1σ of ^{226}Ra and ^{228}Ra in soil, oil sand and sludge samples.

ID	^{226}Ra Bq/kg	^{228}Ra Bq/kg	ID	^{226}Ra Bq/kg	^{228}Ra Bq/kg
Soil			**Soil**		
S1	18.5 ±0.8	22.2 ±2.8	S17	13.5 ±0.7	15.5 ±1.1
S2	22.8 ±1.0	35.5 ±1.9	S20	18.3 ±0.9	19.7 ±1.8
S4	17.4 ±0.6	22.2 ±3.5	S21	11.9 ±0.9	11.1 ±1.0
S6	16.8 ±0.6	17.2 ±1.4	**Oil sand**		
S7	16.5 ±1.3	20.7 ±2.2	OS22-1	22.2 ±0.6	22.4 ±1.8
S8	15.0 ±0.6	15.7 ±1.3	OS22-2	23.5 ±1.4	25.2 ±3.2
S9	13.7 ±1.8	14.7 ±1.3	OS22-3	24.6 ±1.3	25.0 ±1.5
S10	16.6 ±0.6	18.0 ±2.7	OS22-4	24.4 ±0.7	25.3 ±1.6
S11-1	17.3 ±0.8	20.1 ±1.7	OS22-5	25.0 ±1.1	26.0 ±1.4
S11-2	16.8 ±0.6	16.6 ±2.1	OS23-1	20.6 ±0.6	19.3 ±2.0
S11-3	19.2 ±0.6	23.1 ±2.5	OS23-2	20.4 ±0.7	20.4 ±1.4
S11-4	16.1 ±0.9	18.3 ±1.2	OS23-3	22.4 ±1.8	22.9 ±1.5
S11-5	16.5 ±0.8	16.0 ±3.0	OS23-4	21.7 ±1.0	23.3 ±1.4
S12	17.5 ±0.6	19.7 ±2.2	OS23-5	20.7 ±0.7	21.6 ±1.4
S13	18.7 ±0.6	23.5 ±1.3	**Sludge**		
S14	17.3 ±0.6	17.4 ±1.3	SL3	17.8 ±0.7	25.6 ±1.7
S15	18.2 ±0.6	19.7 ±1.3	SL5	22.8 ±0.8	22.9 ±3.6
S16	18.1 ±0.6	17.7 ±1.2	SL19	15.4 ±1.2	17.2 ±1.4

In **Table 2** the average activity concentrations ±1σ standard deviation and the range are summarized for all studied radionuclides. Produced water and crude oil are not reported since all radionuclides measurements correspond to minimum detectable activities (MDA), equal to 0.4, 1.1, 0.4, 1.4, and 0.2 Bq/kg respectively for ^{26}Ra, ^{228}Ra, ^{228}Th, ^{40}K and ^{137}Cs. The results for soil samples are found to be lower or comparable to the world median activity concentrations of ^{40}K, ^{238}U and ^{232}Th, which are respectively 400 Bq/kg, 35 Bq/kg and 30 Bq/kg [19]. ^{137}Cs was detected only in soil and sludge samples, showing high variable concentrations. Indeed, for undisturbed soils with high presence of minerals, ^{137}Cs vertical migration is rather slow and the greater fraction of activity concentration is expected in top soil (0-10 cm). The non-presence of ^{137}Cs in oil sands can be used as a tracer for discriminating oil sand from soil samples. On the other hand, oil sand samples show a higher activity concentration of ^{40}K and ^{226}Ra with respect to soil samples. These concentrations are reasonable considering that oil traps in the Kuçova oilfield are commonly shales, which are known to have abundances of uranium and thorium. Furthermore, it is known that shales contain high abundances of ilmenite and K-feldspar, and therefore high abundances of K.

Table 2. Average activity concentrations (±1σ) and ranges for ^{40}K, ^{226}Ra, ^{228}Ra, ^{228}Th and ^{137}Cs in soil, oil sand and sludge samples.

Sample type	N°	^{40}K Bq/kg	^{226}Ra Bq/kg	^{228}Ra Bq/kg	^{228}Th Bq/kg	^{137}Cs Bq/kg
Soil	21	**297 ±48** 206÷384	**17 ±2** 12÷23	**19 ±5** 11÷36	**20 ±5** 11÷35	**6 ±4** 1÷13
Oil sand	10	**549 ±12** 532÷562	**23 ±2** 20÷25	**23 ±2** 19÷26	**24 ±3** 21÷29	n.d.
Sludge	3	**348 ±115** 220÷443	**19 ±4** 15÷23	**22 ±4** 17÷26	**23 ±6** 17÷28	**7 ±6** 2÷14

n.d. states for "not detected"

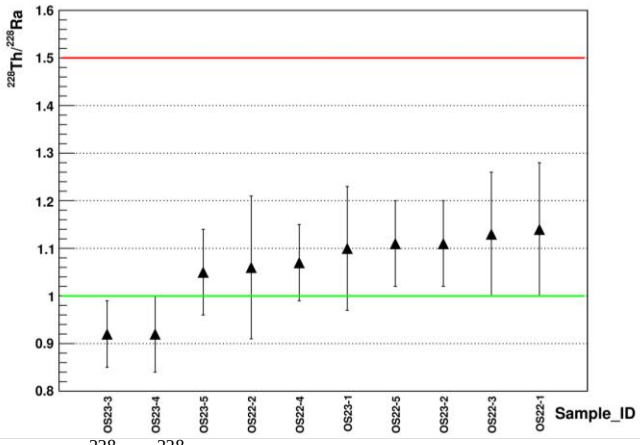

Fig. 3. ^{228}Th/^{228}Ra activity concentration ratios for oil sand samples. The equilibrium condition, corresponding to the case in which both radionuclides have the same activity, is shown with the green line and the maximum ratio assuming transient equilibrium with the red line.

However, radium isotopes are generally unsupported in formation water and, since during oil extraction a fraction of them can precipitate in oil sands, an enhancement of concentration, as well as disequilibrium in the decay chain can occur. Disequilibrium in ^{228}Ra decay segment was studied in terms of the ratio between activity concentrations of ^{228}Th and ^{228}Ra (**Fig. 3**). We observe ratio values systematically greater than unity: however this is

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not a strong evidence of disequilibrium within the standard uncertainties.

3.2 Radiological assessment

The absorbed dose rate (*D*) in air from external gamma radiation at 1 m above ground level due to the presence of uniformly distributed natural radionuclides in measured soils is calculated according to **UNSCEAR** [19]:

$$D\ (\text{nGy/h}) = 0.0417A_K + 0.462A_U + 0.604A_{Th} \quad (1)$$

where A_K, A_U, A_{Th} are the activity concentrations (in Bq/kg) of ^{40}K, ^{238}U (as ^{226}Ra) and ^{232}Th (as ^{228}Ra). Secular equilibrium was assumed for the dose calculation. The average (at ±1σ uncertainty) outdoor absorbed dose rate in the Kuçova oilfield area is 32 ±5 nGy/h. The maximum and minimum calculated values are 21 and 46 nGy/h. This dose is lower than the population weighted average absorbed dose rate in outdoor air from terrestrial gamma radiation (60 nGy/h) [19]. Radiation exposure of workers in the oil-gas industry can occur by increased gamma dose rates due to the relatively higher radioactivity content in oil sand residues. Assuming that oil sands are uniformly distributed over the top soil, the absorbed dose rate was estimated to increase on the average of about 50%, i.e. to increase up to a maximum value of 47 ±5 nGy/h.

The radiological hazard regarding workers and population living in the Kuçova oilfield area is evaluated in terms of annual effective dose rate (*AEDR*). The evaluation of the annual effective dose rate was performed adopting an outdoor time occupancy factor equal to 20% and a conversion factor of 0.7 (Sv/Gy), which accounts for the dose biological effectiveness in causing damage to human tissue.

$$AEDR\ (\text{mSv/yr}) = D \times 10^{-6}\ (\text{mGy/h}) \times 8760\ (\text{h/yr}) \times 0.7\ (\text{Sv/Gy}) \times 0.2 \quad (2)$$

In **Table 3** the results concerning the radiological assessment are summarized. The obtained annual outdoor effective dose rate of 0.04 ±0.01 mSv/y in soils is lower than the worldwide annual effective dose value of 0.07 mSv/y reported by [19]. The excess of dose due to eventual systematic dispersion of oil sand residues in this area, assuming uniform distribution, would lead to an increase of annual effective dose rate of approximately 0.02 ±0.01 mSv/y. This value is negligible considering the recommended limit of excess of effective dose for the population (1 mSv/y) and for workers (6-20 mSv/y).

Table 3. The average (±1σ) and the range of absorbed dose rates (*D*) and annual effective dose rate (*AEDR*) for soil, sludge and oil sand samples.

Sample type	N°	D nGy/h	AEDR mSv/yr
Soil	21	**32 ±5** 21÷46	**0.04 ±0.01** 0.03÷0.06
Sludge	3	**36 ±8** 27÷42	**0.04 ±0.01** 0.03÷0.05
Oil sand	10	**47 ±2** 45÷50	**0.06 ±0.01** 0.05÷0.06

4 Conclusion

The results of this study are the first ever attempt on identifying industrial processes in Albania that may involve the enrichment of NORMs. In this framework, the Kuçova oilfield is chosen as study pilot area, where recently secondary recovery techniques of oil extraction have been introduced. The results will reveal the baseline information for local environmental legacy policies. The activity concentrations of ^{40}K, ^{226}Ra, ^{228}Ra, ^{228}Th and ^{137}Cs in soil samples collected in the Kuçova oilfield are 297 ± 48 Bq/kg, 17 ± 2 Bq/kg, 19 ± 5 Bq/kg, 20 ± 5 Bq/kg and 6 ± 4 Bq/kg, respectively. The ^{40}K and ^{226}Ra activity concentrations in oil sand samples are approximately 85% and 35% higher with respect to values measured in soil samples. These anomalies may be related to the presence of shale traps which are the oil reservoir. However the excess of dose due to eventual systematic dispersion of oil sand residues in this area, assuming uniform distribution, would lead to a negligible increase of annual effective dose rate of population and workers. From a radiological point of view, nowadays, processes of oil extraction do present risks neither for worker nor for the environment.

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