

## **CHARACTERIZATION OF NORM CONTAMINATED SITES AT THE SYRIAN OILFIELD: SAMPLING, ANALYSIS AND DATA MANAGEMENT**

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### **ABSTRACT**

NORM contaminated sites at some Syrian oilfields have been characterized as a part of a national remediation project. Radioactivity, mainly radium isotopes, distributions in surface, subsurface contaminated soil have been determined; volumes of contaminated soil with NORM that need treatment as radioactive wastes, according to the Syrian criteria for clean-up and disposal being evaluated accordingly. Radium isotopes were also determined in ground water of observation wells drilled in the contaminated sites and found to be very low. In addition, radon, which is considered to be a risk arising from contaminated soil by  $^{226}\text{Ra}$ , has been determined in air of subsurface soil. Radon exhalation rates results have been used to determine the cover required to reduce this risk. Moreover, the obtained data was used to carry out Quantified Risk Assessment study and to approach the option for final disposal of contaminated soil.

### **INTRODUCTION**

The occurrence of naturally occurring radioactive materials (NORM) in oil and gas streams has raised the concerns for public, environmental authorities and oil companies (1-7,10,11,12). Production water containing mainly radium isotopes is usually separated from oil and disposed of by down an injection well and disposal wells (8,12,15); all operating oil companies in Syria are currently using this approved disposal method. In the past, uncontrolled disposal of production water has been practiced for several years in all Syrian oilfields that are located approximately 700 km to the northeast of Damascus and near to the City of Der Ezzor. Production water has been collected in unlined artificial lagoons for evaporation; some run-off channels were built to allow water to run-off into the desert. Lagoons and channels become highly contaminated with NORM and could lead to environmental pollution and to radiation exposure of members of the public. This radioactive contamination problem has urged the operating oil companies in Syria to initiate a remediation program in cooperation with the Atomic Energy Commission of Syria for the following purposes:

1. Minimizing risks to environment; particularly those related to ground water.
2. Minimizing risks to oil companies workers and local inhabitants.
3. Minimizing or eliminating existing and potential future environmental liabilities.
4. Complying with the Syrian regulations and international standards.

Therefore, the Atomic Energy Commission of Syria has planned, prepared and initiated a program in cooperation with oil companies. One of the main parts of the program is site characterization. This characterization is considered to be methodic process used to determine the extent and nature of the contamination as well as important site parameters (16). This characterization could include, but not limited, the followings:

1. Determination of radioactivity distribution which is mainly radium isotopes in all environments on site (air, soil, water).
2. Determination of chemical and physical characteristics of the contaminated soil that is considered as radioactive waste and need to be disposed.

Other studies for site characterization were also carried out by the oil companies which include unsaturated zone studies (geological and hydro geological), piezometric studies on ground water, chemical analysis of ground water, litho logical sections determination and others.

The present paper describes only the methods and measurements used to define the contaminated sites from radioactivity point of view. These measurements have been used for the following purposes:

1. Determination of volumes of contaminated soil to be removed to the disposal site according to the Syrian criteria for clean-up and disposal of contaminated soil with NORM (16) and hence the size, design and capping materials for the disposal mounds.
2. The results were used for Quantified Risk Assessment (QRA) study in order to evaluate disposal options (14).
3. Cost Benefit Analysis study, which was carried out by the oil companies.
4. The results to be used as a base line for future environmental monitoring of the disposal site including radioactivity monitoring in groundwater.

## **RADIOACTIVITY DISTRIBUTION IN SOIL**

Detailed site gamma survey and soil characterization was first performed to assess the volume and type of waste materials at all NORM contaminated sites. Gamma survey of the affected areas in all fields was carried out using portable monitors, viz. Ebrline ESP1, Ebrline ESP2 and Saphymo SPP2 NF (counts per second). Radiation contour maps were then established for each site and different zones being defined using Surface Mapping System Software (version 5). This was carried out only to evaluate the extent of contamination and define the sampling strategy.

### **Soil Sampling and Radioactivity Determination**

Soil samples were collected using a stainless steel core sampler with a 7-cm diameter. Only the top 0 – 5 cm layers of surface soil were considered as surface samples. Number of samples taken depends on the area of survey and the used grid for each site. In addition, core samples were collected using the same Auger. The number of core samples depends on the area of contaminated land and the number of the hot spots in each lagoon; more than 3000 surface soil samples and about 200 core samples being collected. In addition, some soil samples from each lagoon were particle size analyzed using automatic mesh shaker.

All soil samples were dried in the oven for 48 hours at 105 °C. The samples were then grinded and about 40g of each sample was filled in a special counting container and stored for two weeks for counting. <sup>226</sup>Ra activities in the collected samples were determined by measuring its gamma emitting daughters, <sup>214</sup>Bi and <sup>214</sup>Pb. Counting was carried out using five gamma

spectrometry systems viz., high-resolution (2 KeV at 1.332 MeV) germanium lithium detector with relative counting efficiency of 10% (Canberra), high-resolution (2.15 keV at 1.332 MeV)

high pure germanium detector with relative efficiency of 25% (PGT), high-resolution (1.7 keV at 1.332 MeV) high purity germanium detector with relative efficiency of 20% (Ortic), high-resolution (1.85 keV at 1.332 MeV) high purity germanium detector with relative efficiency of 46% (Eurisys) and high-resolution (1.85 keV at 1.332 MeV) high purity germanium detector with relative efficiency of 15% (Canberra).

### Radioactivity Distribution

Surface  $^{226}\text{Ra}$  activity distribution in all contaminated lagoons and channels were determined, surface distributions were plotted using Surface Mapping System Software (version 5). The area of contaminated land in each lagoon has been calculated according to the cleanup and disposal of NORM contaminated soil criteria, (soil containing  $^{226}\text{Ra}$  concentration higher than 5.2 Bq/g should be treated as radioactive waste). The highest surface  $^{226}\text{Ra}$ -activity found in the Syrian oilfield was about 100 Bq/g. However, estimation of total volume of contaminated soil in each area was conducted using depth profiles of each lagoon; Figure 1 shows the trend of  $^{226}\text{Ra}$  distribution with depth in one of the contaminated lagoons. The average contamination depth which needs to be treated as radioactive waste in most areas was found to be 50 cm.

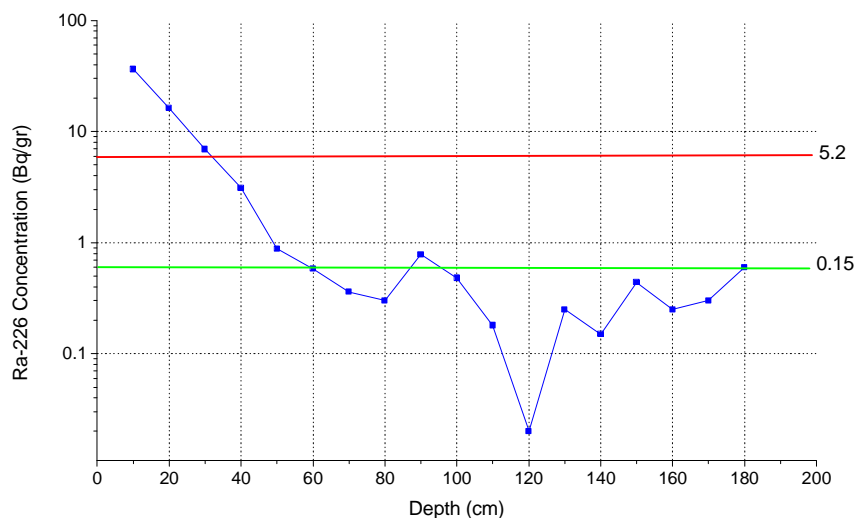


Fig. 1. Depth distribution of  $^{226}\text{Ra}$  in one of the core samples.

### Radioactivity Distribution with Soil Particle Size

Radium-226 was also determined in different particle size soil samples from the contaminated lagoons. Results of some samples are present in Table 1. It was found that radium was only more concentrated in smaller particle than larger ones in one site (TH), but this trend was not observed for all sites where big fluctuations being observed in other samples. However, determination of radium isotopes distribution with particle size is more important for volume reduction of radioactive waste where heavy duty equipment (big sieves) for separation of fine

particles from aggregate material components, high pressure hot water spray is usually used to wash NORM fines (17). In addition, particles size distribution results have also been used for leaching and erosion studies.

Table 1. Radium-226 Distribution with Soil Particle Size of Some Soil Samples Collected from Contaminated Lagoons.

Sample Code	Particle Size (mesh)	Percentage (%)	<sup>226</sup> Ra (Bq/kg)
OM	30	82	14790±520
	170	3	±
	200	5	±
	350	10	±
TH	30		31860+970
	170	5	48980±2040
	200	8	67640±1940
	350	9	90050±3870
AL	30		8910±320
	170	5	8410±250
	200	7	6560±340
	350	8	8930±390

## RADON LEVELS

Radon, the gaseous decay daughter of <sup>226</sup>Ra, presents a health hazard to humans. It can emanate from contaminated soil with <sup>226</sup>Ra and diffuse to the atmosphere. Many regulatory organizations in the world have used radon concentration in air or radon exhalation rates as criteria for cleanup of contaminated land with <sup>226</sup>Ra (18,19). In addition, capping of contaminated soil with clean and suitable cover is one of the methods, which can be used to reduce radon risk. The effectiveness of earthen covers for radon attenuation depends on a variety of factors including the thickness, density and moisture content of the cover and the contaminated soil, radium content and radon exhalation rate. Radon exhalation rate depends also on <sup>226</sup>Ra concentration in soil, soil parameters such as porosity, moisture content, etc., bulk density and the thickness of contaminated soil (19). Therefore, radon concentrations in air and radon exhalation rates have been determined in most of the contaminated sites.

### Radon Measurement in Air

Two nuclear techniques were used for radon measurements, viz. passive and active. The passive technique ("Can" technique) is simply based on digging a hole in the ground (using an auger) of about 11 cm in diameter with a depth of 30 cm. A 50-cm PVC tube is inserted into the hole. At the bottom of the tube, a CR-39 detector attached to the closed top of an inverted plastic container (7-cm diameter and 10 cm height) placed at the bottom of the tube. All containers were left for about 21 days. While the active method is based on sampling of radon gas into a scintillation

chamber coated from inside with zinc sulfide (ZnS), "Lucas Cell". Scintillation produced by alpha particles from radon and its daughters in the closed chamber are counted. Radon inside the cell is allowed to reach secular equilibrium with its daughter products (a waiting time of about 2.5 hours is required). The cell is then coupled to a counting system for measurement. The counting period is typically around 1000 second as it should be small comparable with the half-life of radon.

Relatively higher concentrations of radon in air (up to 81 Bq/m<sup>3</sup>) than the background levels (21 Bq/m<sup>3</sup>) were observed in the surrounding areas of the lagoons. However, these values are lower than the action levels set by NRBP (200 Bq/m<sup>3</sup>) (20). In addition, radon concentrations in contaminated soil with NORM were found to be high. The highest value was found to be about 88134 Bq/m<sup>3</sup>. These high concentrations were in correlation with the high concentrations of <sup>226</sup>Ra present in the contaminated soil.

### Measuring Radon Exhalation Rate

Radon exhalation rate from soil was determined using suitable sized container (1.2-liter) which is placed on the surface of the contaminated soil with its open-end place over the surface. This container is left onsite and radon concentration within the container is subsequently measured at specific time intervals. Radon concentration within the container is measured using the scintillation method (Lucas cell). The results showed that radon exhalation rates in the contaminated lagoons and channels vary between 0.087 Bq m<sup>-2</sup> s<sup>-1</sup> and 12.4 Bq m<sup>-2</sup> s<sup>-1</sup>, all values were higher than the global background (0.01 to 0.05 Bq m<sup>-2</sup>s<sup>-1</sup>) reported by international organization; Table 2 shows some results for site AL. In addition, some values were higher than the action levels (0.74 Bq m<sup>-2</sup> s<sup>-1</sup>) set by Environment Protection Agency (EPA). However, the measured values were used to determine type and thickness of the cover needed to reduce the radon risk.

Table 2. Radon Exhalation Rates (ELW oilfield)

Sampling Location	Gamma Exposure (CPS)	Radon Exhalation Rate (mBq m <sup>-2</sup> .s <sup>-1</sup> )
4	5000	1465
5	4000	967
16	350	2293
17	1700	1537
18	2000	947
19	800	4036
20	1500	7471
21	130	1036
22	800	9137

### **SITE GROUND WATER ANALYSIS**

Radioactive depth profiles results showed that radioactive contamination has reached a depth of more than one meter. In addition, ground water levels in some contaminated areas are rather shallow and water can be seen at a depth of 5 meters. Therefore, leaching of activity from the upper layer of soil may occur; agricultural activities around the contaminated areas are also observed. However, to obtain assurance that NORM contamination is not posing risk to third parties, a sampling and monitoring campaign of water sources was required. Several observation wells around and near to the contaminated land were constructed by oil companies. Samples from these observation wells were taken for analysis every three months. Radium isotopes ( $^{224}\text{Ra}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ) were precipitated as barium (radium) sulfate from 5 liters of each sample using barium carrier. The precipitates were then dissolved in 500 ml of EDTA solution ( $0.5 \text{ mole l}^{-1}$ ). The samples were then filled in a counting container and stored for one month for counting. High resolution and high efficiency gamma detectors have been used to determine radium isotopes in water samples. Counting time was set to obtain good statistic.

All analyzed samples were found to contain  $^{226}\text{Ra}$  activity with values lower than the lower limit of detection of the counting systems. These limits are lower than that found in some drinking water analyzed in our laboratory; some of the measured values are even lower than the permissible maximum concentrations for drinking water (21). However, the obtained results are not sufficient to ensure that there is no evidence of ground water contamination with NORM in the area therefore; these wells should be monitored regularly to ensure that NORM does not reach the ground water.

### **CONCLUSION**

NORM contaminated sites in Syrian oil fields have been characterized. The results of radioactivity distribution in surface and subsurface soil become available for volume determination of contaminated soil that need to be treated as radioactive waste according to the Syrian criteria. Radon measurements have also been provided and can be used for cover design of the disposal pit. In addition, radioactivity base line in ground water of the contaminated sites has been established and can be used for future monitoring.

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