NORM measurements in the oil and gas industry in Argentina

A. Canoba, G. Gnoni, W. Truppa

Autoridad Regulatoria Nuclear
Av. Del Libertador 8250, Ciudad de Bs. As. (1429)
Argentina
Email: acanoba@cae.arn.gov.ar

Abstract. The oil and gas industry, which is especially significant in Argentina, is one industry that concentrates natural radionuclides during its processes. The Nuclear Regulatory Authority of Argentina (ARN) carried out a project with the objective of evaluating NORM, mainly in this type of industry. Seven facilities were characterized, three of them related to the gas industry and four related to the oil industry. In all cases, facility-specific dose rates were measured. First, background measurements were made and then a screening survey was carried out to detect values above background. Of the values obtained, 57% were in the background range, 19 % were below 2 μSv/h, 15 % were in the range 2–10 μSv/h and 9% were above 10 μSv/h. Some values were as high as 400 μSv/h. The annual effective doses were estimated to be in the range 0.02–1.6 mSv/a, far below the dose limit for workers (20 mSv/a), but in some cases above the dose limit for the public (1 mSv/a). The radon gas concentration was also measured in gas facilities. The values obtained showed that radon concentrates in the ethane and propane flows. In addition, samples were taken and later analysed by gamma spectrometry, liquid scintillation and fluorimetry in the ARN laboratories. It was confirmed that the main radionuclides involved are ^{226}Ra and ^{228}Ra and that uranium does not migrate into the oil and gas extraction processes. The radium isotope concentrations measured in some samples were above the exemption values established by the International Basic Safety Standards (IAEA Safety Series No. 115). Finally, protective measures to reduce occupational doses in the cleaning and maintenance processes were suggested, as well as for storage of NORM-contaminated items.

1. Introduction

Radioactive materials containing radionuclides of natural origin are known as NORM (naturally occurring radioactive material). Some minerals have significant levels of natural radionuclides that are extracted and processed with other elements. Some industries involve processes that concentrate natural radionuclides and then may cause some risk to people if the exposures are not under control. These naturally radioactive materials that are concentrated by some industries are known as TENORM (technologically enhanced naturally occurring radioactive material). Although there is a conceptual difference between NORM and TENORM, sometimes the term NORM is used to refer to TENORM. TENORM is found in some effluent flows and wastes from some non-nuclear industries, for example in metal residues, scales, sludges, and fluids. These materials, the by-products and the final products from processes may enhance the exposure to workers and members of the public. The most important radioactivity source in TENORM is due to the presence of isotope products of the uranium and thorium decay chains [1–3].

The presence of radioactive materials of natural origin in geologic formations is well known. The materials containing natural radionuclides found in oilfields are typically located in subsurface formations of oil and gas reservoirs created in the Jurassic period. In the oil and gas industry the techniques used in forcing the oil to the surface include recirculation of produced water, which is extracted with the final products. The NORM materials are transported to the surface with this produced water. A decrease in pressure and temperature results in sulphate and carbonate precipitation inside the pipelines and in the internal surfaces of the equipment. The similar chemical behavior of radium and barium produces selective co-precipitation of both elements in scales. It can also be found other products of the uranium and thorium decay chains. The naturally radioactive material which is not present in scales appears in the vessels with the drained water or in sludges. Other radionuclides of interest, particularly in gas equipment, are radon gas and ^{210}Pb, which usually forms a thin cap in the internal surface of processing equipment [4-5].

From the occupational point of view, the main aspects of radiological protection related to scales and sludges are gamma irradiation and internal contamination of workers arising from the maintenance of equipment containing NORM. The Nuclear Regulatory Authority of Argentina (ARN) carried out a project whose objective was the evaluation of NORM, mainly in this type of industry. With this purpose, seven facilities were characterized, three of them related to the gas industry and four to the oil industry. In this work, the results obtained within the companies surveyed are presented, with the aim of evaluating the presence of NORM and the exposure of workers.

2. Facilities description

2.1. Oil facilities

2.1.1. Facility A

The company provides pumping systems for oil and gas extraction processes. This facility performs the assembling of equipment with new or recovered items. The equipment for recycling arrives at a sector called 'discharging' and from there go to the 'disassembling' sector, where the components are washed, recovered and refurbished. The rejected components are returned to the discharging sector to await disposal as waste or selling as scrap.

2.1.2. Facilities B, C and D

These facilities perform services of cleaning, maintenance and inspection of tubing. They are different bases of the same company. In Argentina, the company has seven bases.

The tubes arrive and are classified and stored in the store area until the washing process begins in the washing area. The wastes from the washing process are temporarily stored until they are removed by the service companies. All processes are performed in well ventilated areas. The washing process is carried out in two steps: first the tubes are introduced to a washing container with a mix of water and gasoline at 90°C for 10–15 min. Then an internal and external manual washing with pressurized water is carried out. The water is collected in vessels called APIs. In some facilities, mechanical equipment is also used to remove scales. The solid wastes from the process are collected in two containers located at both ends of the pipe. These wastes are then manually carried to a large container where they are temporarily stored.

2.2. Gas facilities

2.2.1. Facility E

The company separates and fractionates the heavy components of natural gas (LGN) in two facilities: a separation plant and fractionation plant. In the separation plant, the natural gas is received and dried. Then, the heavy components are sent in the liquid state via a 600 km pipeline to the fractionation plant, were ethane, propane, butane and gasoline are separated. In the fractionation plant there are five main areas: reception of the rich component mix, separation of the rich components, ethane reconditioning, storage areas, dispatch and services. The measurements were performed in the final area.

The distillation process is performed in three continuous stages:

(a) A de-ethanizing tower retains ethane at the top;

(b) A depropanizing tower retains propane at the top;

(c) A third tower retains butane at the top and gasoline at the bottom.

Then, ethane is purified and dispatched; propane, butane and gasoline are stored.

2.2.2. Facilities F and G

These two facilities produce ethylene and polyethylene. The ethylene is obtained from ethane. The polyethylene is produced from ethylene. Facility F has been in operation since 1981 and facility G since 2001.

3. Measurements

To determine whether there were areas or equipment contaminated with NORM, various locations were surveyed. The survey locations were selected on the basis of the processes performed in each place, taking into account the origin, function and visual inspection of the different items. In situ dose measurements were performed and samples were taken for analysis at the ARN laboratories.

3.1. In situ measurements

Dose rate measurements were carried out in predetermined areas. The equipment used was:

(a) Scintillation detector (INa)Tl IDENTIFINDER 1.2” × 1.5”

(b) Geiger-Müller detector AUTOMESS 2174

First, background measurements were performed in the surroundings of each facility. Then, in facilities A, E, F and G, measurements were performed in contact, with the locations being selected on the basis of the origin, function (information given by the staff facility) and visual inspection of the elements (sludge presence). If possible, the internal surfaces of the items were also measured (with a probe). The results are summarized in Table 1, while values above 10 µSv/h found in facility F are presented separately in Table 2.

TABLE 1. DOSE RATES MEASURED IN CONTACT AT FACILITIES A, E, F AND G

	Background (µSv/h)	At facility	
		Range (µSv/h)	Number of measurements in range
Facility A	0.20 ± 0.02	Background	9
		<2	5
		2–10	9
		10–20	2
		>20 (28.2 and 30)	2
Facility E	0.10 ± 0.02	Background	7
		<1	6
Facility F	0.15 ± 0.04	Background	9
		<2	11
		2–10	5
		>10	5
Facility G	0.12 ± 0.03	Background	19
		<1	11
		1–3	16

TABLE 2. DOSE RATES EXCEEDING 10 µSv/h AT FACILITY F

	Dose rate at various distances from the surface (µSv/h)		
	Contact	1 m	3 m
P5601 pump	400	20.0	2.0
P5601 suction pump	320	20.0	-
Pipes at 1 m from P5601 pump	110	-	-
Pipes at 2 m from P5601 pump	30	-	-
5601 pipe	22	5.5	-

In facilities B, C and D, dose rate screening was performed in each area, with the objective of detecting dose rate values above background. After that, detailed measurements were performed at those points where values above background were found. The results are summarized in Table 3, while more detailed results for the washing area of facility D are presented in Table 4.

TABLE 3. DOSE RATES MEASURED IN CONTACT AT FACILITIES E, F AND G

	Background dose rate (μSv/h)	Number of measurements above background	Dose rate in contact (μSv/h)
Facility B	0.09 ± 0.01	1	2.2
Facility C	0.11 ± 0.01	0	—
Facility D, store area	0.13 ± 0.01	1	2.8
Facility D, washing area	0.13 ± 0.01	3	1–10
		1	10–20

TABLE 4. MEASUREMENTS IN THE WASHING AREA AT FACILITY D

	Dose rate at various distances from the surface (μSv/h)		
	Contact	1 m	3 m
Washing container	1.0	—	—
Big container	10.0–18.5	3.0	0.90
Waste container 1	1.0–2.8	—	—
Waste container 2	3.8	0.80	—
API vessel	0.10–0.13	—	—

3.2. Laboratory analysis

Samples of scales, sludges and washing effluents at facilities A, B, C and D were analysed at the ARN laboratory. The scales and sludge samples were obtained from items exhibiting dose rates above background. First, the samples were analysed by gamma spectrometry using a Canberra GeHp detector, model GX2518, with 30% efficiency. Then, ^{226}Ra analysis was performed using a radiochemical method based on the co-precipitation of radium with $BaSO_4$ and the measurement of radon gas by liquid scintillation. The uranium concentration was measured by fluorimetry using Jarrel Ash equipment. The results are summarized in Table 5. At facilities E, F and G, radon gas measurements were performed by the Lucas cell method, in which radon samples were collected in cells coated with SZn(Ag) and then measured using a Ludlum 2200 alpha counter. The results are presented in Table 6.

TABLE 5. URANIUM AND RADIUM CONENTRATIONS IN SAMPLES FROM FACILITIES A, B, C AND D.

	Uranium concentration (μg/g)		Radium activity concentration (Bq/g)			
			^{226}Ra		^{228}Ra	
	Minimum	Maximum	Minimum	Maximum	Minimum	Maximum
Facility A	<0.4	1.9 ± 0.8	<0.1	1270 ± 130	115 ± 11	1670 ± 17
Facility B	<10.0	33.0 ± 9.8	<0.0017	26.8 ± 2.7	<0.0011	9.6 ± 0.9
Facility C	<10.0	1.5 ± 0.7	<0.00014	0.07 ± 0.01	<0.00096	0.1 ± 0.01
Facility D	<0.4	<0.7	0.0019 ± 0.0004	18.7 ± 1.8	0.0021 ± 0.0004	65.4 ± 6.5

TABLE 6. RADON CONCENTRATIONS IN GAS STREAMS AT FACILITIES E, F AND G

	Radon gas concentration (Bq/m^3)
Facility E, ethane + CO_2	1841 ± 300
Facility F, tower top (propane 18%, propylene 75%)	337 773 ± 30 000
Facility G, tower top (propane 18%, propylene 75%)	62 572 ± 5000

4. Results

4.1. External exposure

Dose rates above background were detected in tubing containing scale, in miscellaneous items, in containers for material from washing and maintenance processes and in ethane and propane flows. It was found that 57 % of the dose rates were at background levels, 19 % were below 2 μSv/h, 15 % were in the range 2–10 μSv/h and 9% were above 10 μSv/h.

In order to assess the maximum occupational dose that a worker might receive in these facilities, conservative scenarios were established. Occupancies were calculated on the basis of information provided by the facility staff. Homogeneous whole body irradiation was assumed. The maximum dose rates, occupancies and calculated annual effective doses are shown in Table 7.

TABLE 7. RESULTS OF EXTERNAL EXPOSURE ASSESSMENTS

	Items giving rise to dose rates above background	Maximum dose rate (μSv/h)	Annual occupancy (h)	Annual effective dose (mSv)
Facility A	Miscellaneous items, pipes	30	20 (5 min/d, 240 d/a)	0.6
Facility B	Pipes	2.2	25 (5 min/d, 300 d/a)	0.05
Facility C	None	—	—	—
Facility D[a]	Pipes	2.8	25 (5 min/d, 300 d/a)	0.07
	Container with scales, at 1 m	0.8	320	0.26
	Large container, in contact	18.5	25 (5 min/ d, 300 d/a)	0.45
	Large container, at 1 m	3	50 (10 min/ d, 300 d/a)	0.15
Facility E	De-propanizer pump	0.9	20 (5 min/ d, 240 d/a)	0.02
Facility F	Pump 5601	400	4	1.6
Facility G	Pump P93	3.0	4	0.01

[a] It was assumed that the worker was exposed to all sources and therefore received a total annual effective dose of 0.93 mSv.

4.2 Internal contamination

Inhalation and ingestion of radioactive material are exposure pathways that become important during cleaning and maintenance processes, in which workers may be in contact with particulate material, wastes etc. These pathways will be evaluated in the next stage of the investigations.

4.3. Radon gas concentration

It was confirmed from the measurements performed in the gas facilities that radon is concentrated in ethane and propane flows. This is a result of radon having a condensation point between those of propane and ethane and thus following these products in the distillation and cracking flows.

4.4. Sample analyses

The analyses performed by fluorimetry showed that uranium is not concentrated in scales. This reflects the fact that uranium is not mobilized in the oil extraction process. The analyses performed by gamma spectrometry confirmed that the radionuclides involved come from the decay chains of ^{238}U and ^{232}Th. The radionuclides that mainly concentrate in these processes are ^{226}Ra and ^{228}Ra. Some of the radium concentrations measured in scale samples are above the exemption values established in the International Basic Safety Standards (IAEA Safety Series No. 115 [6]), namely10 Bq/g for ^{226}Ra and ^{228}Ra.

5. Conclusions

The dose rates measured at most facility locations were within normal background levels. Some instances of dose rates above background included tubing contaminated with NORM (facilities A, B and D) and the washing area in facility D. The wastes arising from the washing area are stored in each facility until removal by service companies. It is reported that this material may be used in road construction. In the gas facilities E, F and G, some dose rates were above background in the ethane and propane flows.

In oil facilities, an annual effective dose of 0.6 mSv was conservatively estimated from the highest dose rate measured in tubing. In facility D, assuming that a worker may be exposed to additional scenarios, including duties not only in the store area but also in the washing area, the annual effective dose calculated in a conservative way was 0.93 mSv. It is suggested that the doses received by the workers in these areas be optimized by examining the possibilities for reducing the occupancy periods.

With regard to gas facilities, the values measured in facility F were higher than those measured in facility G, owing to greater accumulations of radionuclides in the older facility. The annual effective dose calculated in a conservative way from the highest value measured was 1.6 mSv. Although the time spent by workers in the areas of highest dose rate is short, it is suggested that the presence of workers in these areas be justified and that their doses be optimized by examining the possibilities for reducing the occupancy times.

The results obtained from this investigation may not agree with the results of future investigations, owing to the fact that the contamination of tubing and various other equipment may vary over time. All the annual effective doses are very low in comparison with the dose limit established in the ARN Standards for workers (20 mSv). In the case of facility F, the value exceeded the limit for members of the public (1 mSv) [6]. In order to improve the dose assessment for workers, it would be advisable to perform TLD measurements over a period of three months and to evaluate the inhalation and ingestion pathways, especially during inspection, repair or maintenance activities because of the possibility of aerosol generation.

For those items giving rise to dose rates above background levels it would be important to define suitable storage methods. As some items are sold as scrap, it is advisable to first clean them to reduce the dose rate. In this respect, protective measures to reduce occupational doses in the cleaning and maintenance processes, as well as in the storage of NORM contaminated items, have been proposed, based on international practice [7–8].

This work has confirmed that the main radionuclides found in this type of industry are ^{226}Ra and ^{228}Ra, members of the ^{238}U and ^{232}Th decay chains, respectively. In some cases, the radium activity concentrations measured in scale samples were above the exemption values established in international standards. This work has also confirmed that uranium is not mobilized in the oil extraction process. On the basis of radon gas concentrations measured in gas facilities, this work has confirmed that radon concentrates in ethane and propane flows. The possibility of gas inhalation should be taken into account during inspection, repair or maintenance activities — in normal operations, the gas is confined within the pipes and vessels with no risk to workers.

Finally, it is suggested that the facilities be re-evaluated to determine the buildup of NORM contamination over time.

REFERENCES

[1] HIPKIN, J., PAYTNER, R.A., Radiation Exposures to the Workforce from Naturally Occurring Radioactivity in Industrial Processes, Radiat. Prot. Dosim. **36** 2–4 (1991) 97–100.

[2] PENFOLD, J.S.S., MOBBS, S.F., DEGRANGE, J.-P., SCHNEIDER, T., Establishment of Reference Levels for Regulatory Control of Workplaces where Materials are Processed which contain Enhanced Levels of Naturally-Occurring Radionuclides, Radiation Protection 107, Office for Official Publications of the European Communities, Luxembourg (1999).

[3] REED, G., HOLLAND, B., McARTHUR, A., "Evaluating the real risks of radioactive scale in oil and gas production", Health Safety and Environment (Proc. 1st Int. Conf. The Hague, 1991), Soc. Petroleum Engrs (1991) 549-558.

[4] EUROPEAN COMMISSION, Reference levels for wokplaces processing materials with enhanced levels of naturally occurring radionuclides, Radiation Protection 95, Office for Official Publications of the European Communities, Luxembourg (1999).

[5] SMITH, K.P., WILLIAMS, G.P., TEBES, C.L., Radiological Dose Assessment Related to Management of Naturally Occurring Radioactive Materials Generated by the Petroleum Industry, Environmental Assessment Division, Argonne National Laboratory, Argonne, IL(1996).

[6] FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS, INTERNATIONAL ATOMIC ENERGY AGENCY, INTERNATIONAL LABOUR ORGANISATION, OECD NUCLEAR ENERGY AGENCY, PAN AMERICAN HEALTH ORGANIZATION, WORLD HEALTH ORGANIZATION, International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources, Safety Series No. 115, IAEA, Vienna (1996).

[7] AMERICAN PETROLEUM INSTITUTE, Bulletin on Management of Naturally Occurring Radioactive Materials in Oil & Gas Production, (Bull. E2), API Official Publications, Dallas, TX (1992).

[8] INTERNATIONAL ATOMIC ENERGY AGENCY, Radiation Protection and the Management of Radioactive Waste in the Oil and Gas Industry, Safety Reports Series No. 34, IAEA, Vienna (2003).