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Radon gas distribution in natural gas processing facilities and workplace air environment

M.S. Al-Masri*, R. Shwiekani

Department of Protection and Safety, Atomic Energy Commission of Syria (AECS), Damascus, P.O. Box 6091, Syrian Arab Republic

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Abstract

Evaluation was made of the distribution of radon gas and radiation exposure rates in the four main natural gas treatment facilities in Syria. The results showed that radiation exposure rates at contact of all equipment were within the natural levels $(0.09-0.1 \ \mu\text{Sv} \ h^{-1})$ except for the reflex pumps where a dose rate value of $3 \ \mu\text{Sv} \ h^{-1}$ was recorded. Radon concentrations in Syrian natural gas varied between 15.4 Bq m⁻³ and 1141 Bq m⁻³; natural gas associated with oil production was found to contain higher concentrations than the non-associated natural gas. In addition, radon concentrations were higher in the central processing facilities than the wellheads; these high levels are due to pressurizing and concentrating processes that enhance radon gas and its decay products. Moreover, the lowest ²²²Rn concentration was in the natural gas fraction used for producing sulfur; a value of 80 Bq m⁻³ was observed. On the other hand, maximum radon gas and its decay product concentrations in workplace air environments were found to be relatively high in the gas analysis laboratories; a value of 458 Bq m⁻³ was observed. However, all reported levels in the workplaces in the four main stations were below the action level set by IAEA for chronic exposure situations involving radon, which is 1000 Bq m⁻³.

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1. Introduction

Naturally Occurring Radioactive Materials (NORM) resulting from the ²³²Th and ²³⁸U-series can be concentrated and accumulated in tubing and surface equipment in the form of scale and sludge as a consequence of physical and chemical processes associated with the oil and gas industry (IAEA, 2004; Jonkers et al., 1997; API, 1992). These scales and sludge from oil production and processing contain mainly radium isotopes and their daughters. On the other hand, a fraction of radon gas is released from reservoir sand and precipitated scales during the decay of radium. Radon is transported with natural gas to gas plants where the raw gas is processed to remove impurities and to recover the less volatile hydrocarbons that separate out naturally in pipelines or in separation plants by condensation to form the so-called Natural Gas Liquids (NGL). Generally, NGL includes ethane, propane and heavier fractions such as butane. Radon appears to be concentrated in the ethane and propane stream relative to the natural gas output of the plant.

^{*} Corresponding author. Tel.: +963 112132580; fax: +963 116112289. *E-mail address:* scientific1@aec.org.sy (M.S. Al-Masri).

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This is due to the fact that the radon boiling point falls between those of propane and ethane. Therefore, the build up of radon decay products (²¹⁰Pb, ²¹⁰Bi and ²¹⁰Po) in internal walls of pipes, vessels of propane and ethane circuits is higher than in other parts of gas plants (Kolb and Wojcik, 1987; API, 1992; Drummond et al., 1990; Heaton and Lambley, 1995; IAEA, 2004).

Many studies on the distribution of radon gas in gas fields and processing plants have been carried out (Drummond et al., 1990; Jonkers et al., 1997; Otto, 1989; Hartog et al., 1995; Hamlat et al., 2003; API, 1992; Gray, 1993; Van Netten et al., 1998). Reported ranges of 222 Rn, 210 Pb and 210 Po concentrations in natural gas are 5–200,000 Bq m⁻³, 0.005–0.02 Bq m⁻³ and 0.02–0.08 Bq m⁻³, respectively (Jonkers et al., 1997; IAEA, 2004). In addition, the distribution of Rn and its decay products in domestic gas supply (Bohus et al., 1995), storage caverns (Neznal and Thomas, 1996) and Liquid Petroleum Gas (LPG) rail cars (Bland and Chiu, 1996) were also determined. Other studies analyzed the pathways by which occupational exposures to radon decay products occur in the oil and gas fields and gas processing plants (Gesell, 1975; Summarily and Prichard, 1985; Heaton and Lambley, 1995; Hamlat et al., 2001; IAEA, 2004). Exposure pathways for radon and its decay products include:

- 1. Gamma external exposures due to penetrating gamma radiation from short-lived radon decay products (²¹⁸Po, ²¹⁴Bi, ²¹⁴Pb and ²¹⁸Po) in the product stream or plated out on interior surfaces.
- 2. Inhalation of radon and its short- and long-lived decay products from leaks and venting of natural gas and its products.
- 3. Inhalation of radon and its short-lived decay products from the combustion of natural gas and its products.
- 4. Inhalation of beta and alpha particles emitted from the long-lived decay products (²¹⁰Po and ²¹⁰Pb) that are released in the process of servicing contaminated equipment.

Since 1996, NORM investigations in Syrian oilfields were limited to remediation projects related to contaminated sites at the oilfields (Al-Masri and Suman, 2003; Othman and Al-Masri, 2004), characterization of scales produced by cleaning operations of equipment and tubulars using high-pressure water jetting (Al-Masri and Aba, 2005) and production water (Al-Masri, 2006). There was no attempt to determine radon concentration in natural gas produced either from the oil wells as associated gas or from the gas wells as non-associated gas. Therefore, the present paper describes the radon gas distribution in different natural gas treatment units of the main four natural gas treatment plants in Syria. Radon gas concentration in the workplace air environment due to leaks, venting and other sources were also estimated.

2. Materials and methods

Four main natural gas treatment facilities and their related substations were studied. These include Dier Ezzor Directorate, Al-Jbissa Directorate, Al-Hassaka Directorate and the Middle Region Directorate, Fig. 1. Table 1 summarizes production date, substations, type of natural gas and natural gas processing by-products. All natural gas treatment facilities at all directorates have similar specifications.

2.1. Gamma survey of gas installations

The levels of contamination on the equipment and pipelines were measured by holding the scintillation probe (Saphymo SPP2 NE) and a dose rate meter (Eberline ESP1) close to the equipment wall at zero distance. Background levels were measured in each location well away from the location where contamination was expected, using clean new vessels or pipes; measurements showed background values in the average of $0.09-0.1 \,\mu\text{Sv}\,\text{h}^{-1}$ as dose rate and 25–30 CPS as a contamination reading.

2.2. Determination of radon in natural gas

Natural gas sample was collected from the different locations at each gas processing facility, wellhead and substation using a special pressurized cylinder following the method developed by Shell International Research Maatschappij, B.V. (1995). The gas was then drawn into a Lucas cell through a moisture absorber (Silica–gel) by depressurizing process to get the gas sample inside the Lucas cell with normal pressure. After 3 h, radon concentration inside the Lucas cell was determined by counting the scintillations produced by alpha particles from radon and its decay products using a counting system (AB-5 pylon). Radon concentration was then calculated using the following equation:



$$\operatorname{Rn}\left(\operatorname{Bq} \mathrm{m}^{-3}\right) = \frac{C_{\mathrm{S}} - C_{\mathrm{Bg}}}{E \frac{A}{C} v} 1000 \tag{1}$$

where $C_{\rm S}$ is the counts per minute for the sample, $C_{\rm Bg}$ is the counts per minute for background. *E* is the efficiency of the system determined for each cell. This factor is typically 100–150 CPM/Bq. *C* is the radon correction factor for decay during counting. *A* is the radon correction factor for decay of radon from time of collection to start of counting. *v* is the volume of counting cell (1).

| Table 1 | |
|--|--|
| Natural gas fields and treatment plants in Syria | |

| Directorate | Capacity (Mm ³) | Production date | No. of substation | Type of raw natural gas | Products | Operating companies |
|---------------|--------------------------------|-----------------|-------------------|-------------------------------|------------------------------|------------------------|
| Dier Ezzor | 13 | 2001 | 9 | Associated and non-associated | PNG, LPG, sulfur, condensate | DEZPC and AFPC and SPC |
| Al-Hassaka | 0.66 | 1985 | 4 | Associated | PNG, LPG, sulfur, condensate | SPC |
| Al-Jbissa | 2.98 | 1988 | 4 | Associated | PNG, LPG, sulfur, condensate | SPC |
| Middle region | - | 1993 | 4 | Non-associated | PNG, LPG, condensate | SGC, HPC |

LPG, liquid petroleum gas; PNG, pure natural gas; DEZPC, Dier Ezzor petroleum company; AFPC, Al-Furat petroleum company; SPC, Syrian petroleum company; SGC, Syrian gas company; HPC, Hayan petroleum company.

2.3. Determination of radon and its decay products in workplace air environment

Radon concentrations in different places inside the studied gas stations were measured using the solid-state nuclear track detectors (CR-39) method (Durrani and Bull, 1987). About 34 detectors were distributed and left for 3 months at different places of the four gas treatment plants and their substations, such as offices, accommodation rooms, control rooms, compressors houses, laboratories, worker rest rooms and restaurant. The detectors were then collected and the tracks were developed by chemical etching and counted using an optical microscope. From the average track densities on the detectors, radon concentrations were determined.

Radon decay product concentrations were determined as follows: air samples were drawn through a filter using a rechargeable pump for 10 min with a flow rate of $10 \, l \, min^{-1}$. The filter was then removed and placed on a ZnS(ng) scintillation detector and counted using the AB-5 pylon (IAEA, 1976).

3. Results and discussion

Table 2

3.1. External gamma radiation surveys

Gamma radiation emitted from radon decay products (214 Bi, 214 Pb and 218 Po) at the outer walls of the equipment and pipes of all installations was found to be within the background levels (0.09–0.1 µSv h⁻¹) except for two locations at the reflux pumps of Al-Hassaka and Dier Ezzor Directorates, Table 2. The highest dose rate and contamination reading were 3 µSv h⁻¹ and 2000 CPS, respectively. These levels are due to accumulation of short-lived radon decay products (214 Bi, 214 Pb and 218 Po) onto the internal walls of pipes and components of the reflux pumps. These shortlived radon decay products emit high-energy gamma radiation so that they can be detected near the external surfaces (Kolb and Wojcik, 1987; API, 1992; Drummond et al., 1990; Heaton and Lambley, 1995; IAEA, 2004). The levels of gamma radiation decline very quickly with time to the background level when the pumps were switched off, due to the decay of these short-lived decay products. However, the build up of radon decay products (210 Pb, 210 Pb and 210 Po) in the internal walls of pipes and pump surfaces can be seen when measurements are carried out on the internal surfaces.

3.2. Radon distribution in Syrian natural gas and its by-products

Radon was measured at different locations of the Syrian oil and gas fields. Two types of raw natural gas have been collected and analyzed: Associated (ANG) and Non-Associated Natural Gas (NANG). In addition, Pure Natural Gas (PNG) (dry natural gas "pipeline quality"), Liquid Petroleum Gas (LPG) and Sulfur containing Natural Gas (SNG) have been also measured. Radon concentration varied between 15.4 Bq m⁻³ and 1141 Bq m⁻³ (Table 3). These levels are within the reported worldwide range of 5-200,000 Bq m⁻³ (Jonkers et al., 1997). In general, natural gas associated with oil production was found to contain higher concentrations than the non-associated natural gas. These relatively high levels in associated natural gas may be due to the fact that radon in associated gas is generated from different sources: free gas from reservoir with the natural gas, degassing from oil containing dissolved radon and

| Gamma radiation survey results | | | | | |
|--------------------------------|---|----------------------------|---|--|--|
| Directorate | Unit or equipment | Range of gamma count (CPS) | Range of radiation dose $(\mu Sv h^{-1})$ | | |
| Dier Ezzor | Water tank, NLG, manifolds, separators units, substations, gas wellheads, condensation, storage tanks, heat treatment units, pipelines | 25-30 | 0.09-0.1 | | |
| | LPG metering | 50-60 | 0.09-0.1 | | |
| | Reflux pumps | 800-1750 | 2.5-3.00 | | |
| Al-Hassaka | Manifolds, LPG units, distillation units, propone unit, condensation unit and storage tanks, sulfur unit, compressors, fractionation unit | 25-30 | 0.09-0.1 | | |
| Al-Jbissa | Pressure regulators units, sweeting unit, H ₂ S separation unit, dehydration unit, LPG unit, storage and filling units, sulfur unite, substation | 25-30 | 0.09-0.1 | | |
| | Reflux pumps | 1800-2000 | 2.5-3.0 | | |
| Middle region | Manifolds, separators units, compressors, metering units, substations | 25-35 | 0.09-0.1 | | |

Table 3 Distribution of Rn in Syrian natural gas and its products

| Directorate | Sample type | Sampling location | 222 Rn concentration (Bq m ⁻³) | |
|---------------|-------------|------------------------|---|--|
| Dier Ezzor | NANG | Well202 | 55 ± 4 | |
| | NANG | Well203 | 74 ± 3 | |
| | NANG | Well206 | 53 ± 7 | |
| | NANG | Well207 | 15 ± 2 | |
| | PNG | Export line | 264 ± 5 | |
| | ANG | Thaaban substation | 532 ± 4 | |
| | ANG | Al-Jafra substation | 281 ± 7 | |
| | NANG | Processing facility | 78 ± 11 | |
| | ANG | Processing facility | 382 ± 2 | |
| | ANG | Al-Galban substation | 530 ± 7 | |
| | ANG | Al-Kaahar substation | 292 ± 3 | |
| | ANG | Al-Maleh substation | 1142 ± 20 | |
| | LPG | LPG unit | 189 ± 14 | |
| Al-Hassaka | LPG | LPG unit | 225 ± 12 | |
| | PNG | Facility outlet | 462 ± 81 | |
| | PNG | Factory lines | 317 ± 51 | |
| | NANG | Drying substation | 321 ± 73 | |
| | ANG | Swedea 1 substation | 251 ± 33 | |
| | ANG | Swedea 2 substation | 449 ± 26 | |
| | ANG | Swedea 3 substation | 1027 ± 118 | |
| | ANG | Karachok substation | 509 ± 103 | |
| | SNG | Sulfur plant | 64 ± 18 | |
| Al-Jbissa | LPG | LPG unit | 659 ± 119 | |
| | PNG | High-pressure gas | 182 ± 16 | |
| | PNG | Facility outlet | 498 ± 42 | |
| | PNG | Factory lines | 566 ± 67 | |
| | ANG | Ghona substation | 443 ± 21 | |
| | ANG | kebebeh substation | 937 ± 99 | |
| | ANG | Markheda substation | 448 ± 108 | |
| | ANG | Reflux pump | 471 ± 35 | |
| | SNG | Sulfur plant | 80 ± 41 | |
| Middle region | NANG | Najeeb substation | 316 ± 64 | |
| | NANG | ARAK substation | 507 ± 128 | |
| | NANG | Al-Suknaa substation | 133 ± 6 | |
| | NANG | Al-Dubiat substation | 284 ± 130 | |
| | NANG | Al-Heil substation | 330 ± 57 | |
| | PNG | Directorate substation | 459 ± 98 | |
| | PNG | ARAK substation | 298 ± 70 | |
| | NANG | Al-Dubiat substation | 675 ± 167 | |
| | NANG | Al-Hail substation | 686 ± 82 | |
| | NANG | Well 20 | 145 ± 32 | |

NANG, non-associated natural gas; PNG, pure natural gas; ANG, associated natural gas; LPG, liquid petroleum gas; SNG, natural gas containing sulfur.

exhalation from scales containing ²²⁶Ra and precipitated onto the inner surfaces of pipes and equipment. In addition, radon concentrations in natural gas collected from the treatment plants were higher than the concentrations at the well-heads; these are due to pressurizing and concentrating processes that enhance radon gas and its decay products. Moreover, at the treatment plant, the lowest ²²²Rn concentration was in the natural gas fraction used for producing sulfur, a value of 80 Bq m⁻³ was observed. Furthermore, the data collected from all locations for ²²²Rn have been statistically evaluated using the so-called Exploratory Data Analysis method (one example of which is the Box Plot used herein) (Fig. 2). Large variations of ²²²Rn concentrations were found in the associated natural gas. These variations are due to variations in radon gas sources in each field; free gas, exhalation of radon from scales and degassing from oil. Smaller variations were observed in the non-associated natural gas streams.

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Fig. 2. Distribution of 222 Rn in natural gas and fractionation products. The box plot shows the median value and the 25% and 75% percentiles. The short horizontal lines indicate the 10% and 90% percentile, while the asterisks represent outlying values (see Table 3 for definition of the location acronyms).

3.3. Radon concentrations in workplace air environment

Radon and its decay products' sources at the gas treatment plants workplaces are mainly due to leaks, venting and accidental release of natural gas. Radon and its decay products (214 Bi, 214 Pb and 218 Po) were only measured in the workplaces where workers are expected to be present. These include offices, control rooms, accommodation rooms, restaurants and compressors houses. Mean and ranges of 222 Rn, 214 Bi, 214 Pb and 218 Po concentrations at these locations are presented in Table 4. The 222 Rn concentration varied between 14 Bq m⁻³ (background level) and 458 Bq m⁻³. The highest values observed were for the gas analysis laboratories (around 6 m × 4 m × 3 m each laboratory) of the four Directorates. Radon decay products concentrations were also high at the same location. These relatively high levels are due to release of radon and its decay products to laboratory air environment by flashing the gas sample during analysis; the laboratories are airtight to prevent any dust entering the laboratories. This exposure pathway to radon and its decay products should be considered. The same measurements were repeated in these laboratories after recommendations not to flash the natural gas samples inside the laboratories and installing good ventilation systems. Results showed that radon concentrations and its decay products are returned back to normal levels (24 Bq m⁻³). However, all reported levels in the workplaces in the four main stations are below the action level for remedial action relating to chronic exposure situations involving radon in workplaces; is a yearly average of 1000 Bq m⁻³ (IAEA, 1996).

4. Conclusion

Radon concentrations and gamma exposures in Syrian natural gas treatment plants and substations have been determined. Gamma dose measurements were within the background levels at all process equipment and pipes of

| Mean and ranges of ²²² Rn, ²¹⁶ Po, ²¹⁴ Bi and ²¹⁴ Pb concentration in indoor air at different locations of gas held | | | | | | |
|---|--------------------|-----------------------------------|---|---|-----------------------------------|--|
| Location | No. of measurement | 222 Rn (Bq m ⁻³) | ²¹⁸ Po (Bq m ⁻³) | ²¹⁴ Bi (Bq m ⁻³) | 214 Pb (Bq m ⁻³) | |
| Compressor house | 8 | 24.8 (15-37) | 18.7 (0.1-38) | 17.0 (8.9-22.0) | 6.9 (0.2-22.9) | |
| Control rooms | 10 | 21.3 (13-33) | 24.8 (1-79.1) | 19.2 (4.3-49.1) | 8.2 (1.1-18.6) | |
| Workers offices and accommodation rooms | 12 | 31.0 (14-58) | 24.8 (7-38) | 10.2 (6-12.7) | 6.9 (3.2–10) | |
| Laboratories | 4 | 153 (24-458) | 144 (31-444) | 144 (20-500) | 105 (4.8-387) | |

Table 4

the treatment plants and substations except for the reflux pumps where high readings exist. Radon concentration in the Syrian natural gas and its products are within the worldwide reported levels. In addition, radon concentration in associated natural gas was higher than in the non-associated natural gas. Regardless of the other reported pathways for radon and its decay products exposure, a new pathway of radon exposure in the gas industry due to releasing natural gas during analysis at the gas laboratories should be considered; precautions to reduce the risk arising from inhalation of radon are needed.

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