

NATURAL RADIOACTIVITY AT PODRAVINA GAS FIELDS

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Abstract

In Croatia, natural gas is an important source of energy, where its use exceeds other sources by one third. Composed primarily of the methane, natural gas from Croatian Podravina gas fields, beside other impurities, contains small amounts of radioactive elements.

At *Gas Treatment Plant* (GTP) Molve, technological procedures for purification of natural gas and its distribution are performed. With yearly natural gas production of $3.5 \cdot 10^9 \text{ m}^3$ GTP Molve is major Croatian energy resource. Its safety and environment impact is matter of concern.

Using different radioactivity measuring techniques the exposure of population to ionizing radiation were calculated at Central Natural Gas Station "Molve", and the underground wells. The measurement techniques included *in-situ* gamma spectrometric measurements, from which contribution to absorbed dose of the natural radionuclide in soil were calculated. Exposure dose measurements were performed using TL-dosimeters, and "ALARA" electronic dosimeters as well as field dose rate meter. Comparing used different radioactivity measuring methods, the correlations have been calculated.

Introduction

Natural gas is a naturally occurring mixture of gaseous hydrocarbons. The approximate composition of natural gas from Croatian Podravina gas fields is 85 % methane, 10 % ethane, 3 % propane, with lesser amounts of butane, and besides other impurities, contains hydrogen sulfide and mercaptanes, as well as small amounts of radioactive elements.

Energy policy in the late 1980s was aimed at reducing dependence on foreign sources and utilizing domestic power resources more fully. For conventional power generation, the power industries began replacing heavy oil with coal in thermoelectric generators and relying more heavily on hydroelectric stations. Development of nuclear power generation was limited by public resistance (especially after the Chernobyl disaster in 1986) and by lack of domestic technology and nuclear fuel.

Natural gas was discovered in 1917 at Bujavica field. Production of natural gas increased significantly in the 1980's with the opening of the Molve field in 1985 and three wells in the northern Adriatic in 1989. Molve increased production by one-third, yielding one billion cubic meters annually.

"Podravina" is the project, which comprises facilities and systems for gas and other hydro-carbonates production, originating from northwest Panon Area of approximately 250 km^2 . About fifty oil/gas wells, five gas stations (Stari Gradac, Kalinovac Istok,

Kalinovac IP, Kalinovac Zapad i Molve Istok) and Central Gas Treatment Plants (GTP) Molve I, Molve II and Molve III, yield annual gas production of 2.5 billions m³[1].

Natural gas is pipelined to GTP Molve. Here, at GTP Molve III, technological procedures for purification of natural gas and its distribution are performed. With yearly natural gas production of 3.5 10⁹ m³ GTP Molve III is major Croatian energy resource. Its safety and environment impact is matter of concern.

Over the past few years the Radiation Protection Unit of the Institute for Medical Research and Occupational Health in Zagreb, Croatia has been investigating natural radioactivity at Podravina gas fields, as the part of more extensive monitoring program “Monitoring okolisa CPS Molve” (“Environmental Monitoring at Central Gas Treatment Plant Molve”) [2,3,4,5]. The purpose of this paper was to compare different radioactivity measuring techniques and the expose of population to ionizing radiation.

Material and Methods

Gas Treatment Plant is about 2 km west from the community of Molve, where lives around 2000 people. The nearest major town is Koprivnica, located about 18 km northwest GTP, where inhabit around 30 000 residents (Fig. 1).



Figure 1. Podravina gas fields with Molve village

The radioactivity measurement program carried out at gas field Molve was established by experts from the Institute, in co-operation with competent assistants from the Gas Treatment Plant, as well as from local County Office for Public Health.

Locations:

- Central Gas Treatment Plant (GTP)
- Active gas well (M-9)
- Closed gas well (M-10)

Active gas well (M-9) is about 500 metre far from GTP, while closed gas well (M-10) is more distant, about 1000 metre. The community of Molve is about 2 km west from GTP (Fig. 2).

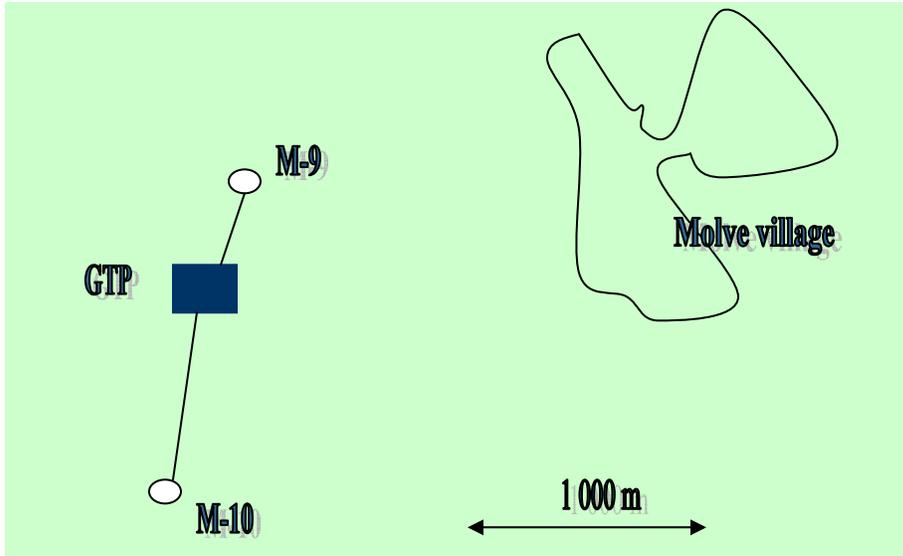


Figure 2. Scheme of Gas Treatment Plant and Molve village

The next measuring methods have been applied:

Measuring methods:

- *In-situ* gamma-spectrometric measurements
 - Continuous measurements of exposure dose rates by use of thermoluminescent dosimeters (TLD)
 - Continuous measurements of exposure dose rates by use of active electronic dosimeters AED “Alara”
 - Direct measurements of dose rate by field instrument
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- *In-situ* gamma-spectrometric measurements have been performed by use of HPGe detector and multi-channel analysing equipment connected with PC. The detectors’ characteristics were: resolution 1.75 keV at 1.33 MeV ⁶⁰Co and 810 eV at 122 keV ⁵⁷Co; relative efficiency 21% at 1.33 MeV ⁶⁰Co; peak-to-Compton ratio 53:1. Measuring time was 1000 seconds on each location.
 - Continuous measurements of exposure dose rates were performed using two instruments:
 - Victoreen TL dosimeters, type TL-33 “Hot Press Chip” (CaF₂:Mn) and reader Victoreen 2008. Dosimeters have been placed at all three locations, exposed over one year - from July 2004 till October 2005.
 - Active Electronic Dosimeter “Alara”; the data have been calculated as absorbed dose rates. Two dosimeters have been joined with Institute’s Unit for Environmental Hygiene equipment (one month in winter period,

and one month during summer, in 2005). One dosimeter was with field team while the environmental radioactivity measuring was performed (from time going out of the Institute, till coming back to the Institute).

- Dose rate measurements were performed using field instrument KOMO TN. On each location measurements were taken momentary for five times, and the mean value was calculated.

Results and Discussion

In-situ gamma-spectrometric measurements have been performed in October 2005. With presumption of equal distribution of natural radionuclides in soil, their concentrations in soil have been calculated. For calculation of ^{137}Cs contribution to absorbed dose, the surface distribution, with exponential penetration in soil was presumption. The contribution to absorbed dose from uranium and thorium natural radioactive series, as well as natural ^{40}K and fission ^{137}Cs have been calculated, and results are presented in Figure 3.

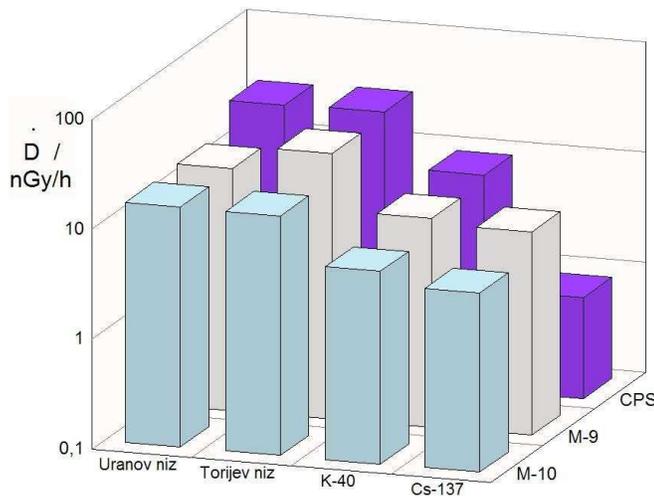


Figure 3. The contribution to absorbed dose from uranium and thorium natural radioactive series, ^{40}K and ^{137}Cs

Radionuclide concentrations from uranium and thorium natural radioactive series, as well as ^{40}K in soil are higher at location GTP than at locations M-9 and M-10, what leads to higher absorbed dose at location GTP.

^{137}Cs surface activity at wells is order of magnitude higher (at M-9 is $2767 \pm 94 \text{ Bqm}^{-3}$; at M-10 is $2022 \pm 82 \text{ Bqm}^{-3}$) than at location GTP ($290 \pm 40 \text{ Bqm}^{-3}$). Being fission radionuclide, ^{137}Cs concentrations are not caused by natural gas production. ^{137}Cs has occurring in the environment since first nuclear experiments, especially after nuclear accidents.

From *in-situ* gamma-spectrometric data equivalent doses (H) were calculated (Table 1).

TL dosimeters have been installed at three locations: GTP, M-9 and M-10. They have been exposed for the period from July 2004 till October 2005 (455 days). During some repairs at location M-9, dosimeter was lost. From exposure dose rates data, the exposure doses per year (X) were obtained, and from these values equivalent doses (H) have been calculated. Insignificantly higher value was measured at GTP, but both values are at the same order of magnitude (Table 1).

Active Electronic Dosimeters AED “Alara” have been installed at the locations M-9 and M-10 during winter and summer periods, each time for one month. One dosimeter was with team all the time during the field exercise. Absorbed dose rates were calculated and results are presented in Figures 4 and 5 respectively. Straight lines present mean values.

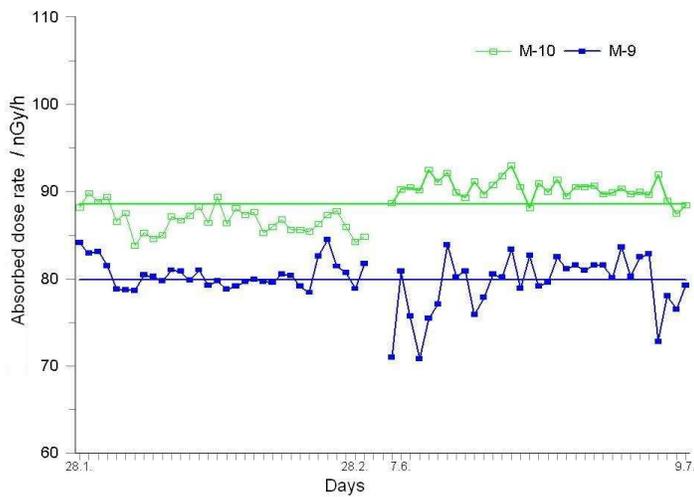


Figure 4. Absorbed dose rates during winter and summer periods

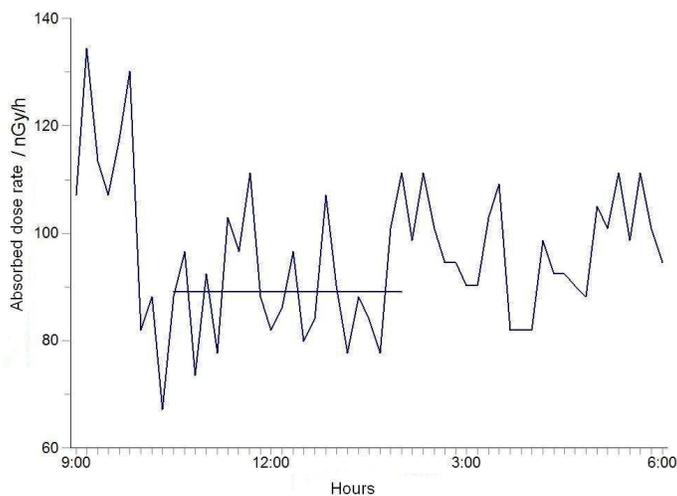


Figure 5. Absorbed dose rates during field measurements

From absorbed dose rates, equivalent doses were calculated and given in Table 1.

Direct measurements dose rates were performed on each location five times, and absorbed dose rate was calculated as the mean value (Table 1).

In Table 1 are presented the results for equivalent doses per year, obtained by different measuring techniques.

Table 1. Equivalent doses obtained by different measuring techniques

LOCATION	DOSES (H / mSv)			
	<i>In-situ</i> gamma spectrometry	TL dosimeters	AED “Alara”	Direct measurements
GTP	0.916	1.16	-	0.96-1.05
M-9	0.710	lost	0.700	0.88-1.05
M-10	0.766	0.97	0.776	0.88-1.05

There is a good agreement between single measuring techniques for each location at gas field Molve. The values do not differ from values obtained in other parts of Croatia; for example, in Zagreb equivalent dose measured in 2004 was 1.22 mSv [6]. Correlation coefficients are from 0.710 (*in-situ* gamma spectrometry-direct measurements) to 0.999 (*in-situ* gamma spectrometry- AED “Alara”) with significance 0.05.

Conclusion

Investigations have shown that natural radioactivity at gas field Molve is not increased compared to other parts of Croatia. Technological process at Gas Treatment Plant has no influence on the environment around the plant. The best proofs are nearby stork’s nests.



References

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