THOROUGH INVESTIGATION OF RADON CONCENTRATION VARIATIONS IN BAITA BIHOR (ROMANIAN NATIONAL RADIOACTIVE WASTE REPOSITORY – DNDR)

A. PETRESCU[1], L. DONE[1], F. DRAGOLICI[1], I. PRISECARU[2], G. PAVEL[2], H. POPA[3]

[1] Horia Hulubei National Institute for Nuclear Physics and Engineering, P.O.Box MG-6, RO-077125 Bucharest-Magurele, Romania, E-mail: alin.petrescu@nipne.ro; donelaur@yahoo.com; fdrag@nipne.ro

[2] University Politehnica of Bucharest, Splaiul Independentei 313, P.O. Box: 060042, Bucharest, Romania, E-mail: gabriel.pavel@gmail.com

[3] Technical University of Civil Engineering Bucharest, Bd. Lacul Tei 124, sector 2, Bucharest, Romania, E-mail: horatiu@utcb.ro

Received March 31, 2014

Radon has an important contribution to natural radiation dose received by population. In some areas Radon contribution to total dose goes up to a level of 95% leading to an exposure to natural radiation much higher than normal. The paper present the investigations conducted in order to evaluate the radiation protection measures applied at Baita Bihor radioactive waste repository in accordance to national and international regulations and to analyze the efficiency of the ventilation system. Radon accumulation in uranium ore mine galleries used as final repository for radioactive waste, such as Baita repository, is a complex issue and deals with certain properties that make it harmful to humans. The study was conducted taking into consideration two different seasons (summer and fall) in order to observe the influence of atmospheric conditions. There were analyzed the variations of radon concentration during four experimental sessions conducted in various environmental conditions. Representative time samples were taken into account, including also specific parameters as operational periods inside repository, the time distribution through the year.

Key words: radon, measurement, radiation protection.

1. INTRODUCTION

Uranium ore contains natural uranium comprising of 99.275% U^{238}, 0.715% U^{235} and 0.005% U^{234}. From radiation protection point of view, U^{238} and its decay products are of major concern for uranium mining industry. The entire spectrum of decay products existing in the ore depends on the age. In general, a secular equilibrium status of the ore is found [1].

External gamma level and inhalation exposure due to radon (Rn^{222}), its short-lived progeny also to the long-lived alpha activity associated with ore dust constitute the major source of radiological hazard in uranium mines.

Monitoring the radon concentration inside uranium mines and in the environment has been a matter of concern since last several decades to minimize the extent of inhalation exposure of occupational workers and the public [2].

Descendants of radon are radioactive solids that adhere to the dust particles in the air which can be inhaled or ingurgitate. If contaminated dust is inhaled or if swallowed, these radioactive elements adhere to internal organs (lungs, digestive tract). As result, an increase in cancer incidence can be observed. The highest exposure to α particle, coming from radon decaying, is undertaken by miners working in badly ventilated uranium mines.

Romanian National Radwaste Repository (DNDR) is located in a complex of galleries built in an ex Uranium Mine used for exploration, situated in Baita-Bihor County, established as disposal of low and intermediate level radioactive waste. It was designed in 1983 and built according to initial project until 1985.

Actual repository is arranged along two main galleries (50 and 53) including their already built transversal pathways.

Underground facilities designed for disposal of radioactive wastes have been used safely for about half a century. Some examples [3] of such repositories are given below: Russian injection (RUS), Richard (CZ), Bratrstvi (CZ), Forsmark (S), Olkiluoto and Loviisa (FIN), WIPP (USA). Among these only Bratrstvi site has a similar design as the Baita Bihor repository. It was built in a former uranium mine and put in operation since 1974. Radon levels are strictly monitored and disposal practices are ceased.

Radon levels can be of great importance not only in the working of mining areas but also in their vicinity. In this sense, a thorough radon survey has been carried out [4–6] in Băita-Stei area, in settlements (*e.g.* Băiţa, Nucet, Fînaţe, Cîmpani) situated in the vicinity of former Avram Iancu uranium mine, aiming to investigate the influence that these waste materials from mines had on different construction (building) types. The authors analyzed the influence of the uranium mines had on in-house radon concentration. Also the authors studied [5, 6] the influence that room types (open space with good ventilation; closed space with poor ventilation) had on radon accumulation.

Extensive radon surveys were conducted in different European countries [8, 9, 10] in order to stress out the importance of ventilation on radon accumulation. In the study [7], the authors investigated the average radon levels in underground workplaces (basements) and the occupational exposure according to national standards. Investigation method was based on long period observation on radon accumulation in about 1000 measuring points.

Impact of ambient temperature and pressure on radon transportation within porous media was also investigated [9] using long-term radon monitoring method. The investigation was focused on radon accumulation and transportation (using alpha and gamma measurement) both in isolated sites (tunnel) from outer meteorological influence and in sites open to the influence of environmental conditions. The results in this case indicate that the diurnal, intra-seasonal and seasonal variations in the radon concentration are clearly associated with the ambient temperature gradient outside the rock air interface.

In order to evaluate the effectiveness of the radioprotection measures in place at disposal working areas from Baita Bihor repository, there were conducted a series of measurements of radon concentration and other parameters (temperature, relative humidity, pressure) inside the Baita-Bihor DNDR galleries.

In this respect and due to the fact that the repository is located in a former exploration uranium mine, the concentration measurement of radon in DNDR galleries is very important for the radioprotection of the operating personnel. The repository was designed and built with a ventilation system which aims to maintain the radon concentration during the disposal operations of radioactive waste packages below the limits imposed by regulations [11]. In terms of the equivalent concentration balance, the effective dose limit for occupationally exposed workers, of 20 mSv/y, is given by a radon concentration of 1110 Bq/m^3.

2. EXPERIMENTAL SESSIONS

The equipment used in the study is SARAD GmbH RTM1688-2. It offers two calculation modes for the Radon concentration, one (Slow) includes both, Po^{218} and Po^{214} decays and the other one includes Po^{218} only (Fast).

The advantage of the “Fast” mode is the quick response to concentration changes while the “Slow” mode gives sensitivity twice as high compared with the fast mode. The higher sensitivity reduces the statistical error of a measurement which depends only on the number of counted decay events.

The measurements of the Radon concentrations performed inside the DNDR Baita-Bihor were carried out during several months and performed in 3 sessions: measurements performed along the transport gallery 50; measurements performed in point 11 of transport gallery 50 (located in front of the galleries 31/1 and 31/2); at the end of the gallery 27/2 (point 12).

First experimental session, associated with an important series of measurements, was conducted in order to determine the precise locations in the disposal area in which radon concentrations are the highest. Usually radon accumulations occur in places inaccessible for natural air currents. Radon concentration was measured in 12 representative points, Figure 1, located at the entrance of the repository, at in the front of the empty or partially filled transverse galleries and in the farthest point of the transport gallery (Gallery 50). Obviously, they are also representative from the radioprotection point of view, being the route for transport, manipulation and disposal of conditioned radwaste packages. Measurement points according to location into galleries are represented in Table 1.

Table 1

Measurement points and their position

Measurement point	Positioning
1	10 m away from the entrance in gallery 50
2	in front of hose 1
3	in front of gallery 9/1 and 9/2
4	in front of gallery 11/2
5	in front of gallery 13/1
6	in front of gallery 15/2
7	in front of gallery 17/2
8	in front of gallery 53
9	in front of gallery 23/1 and 23/2
10	in front of gallery 27/1 and 27/2
11	in front of gallery 31/1 and 31/2
12	at the end of gallery 27/2

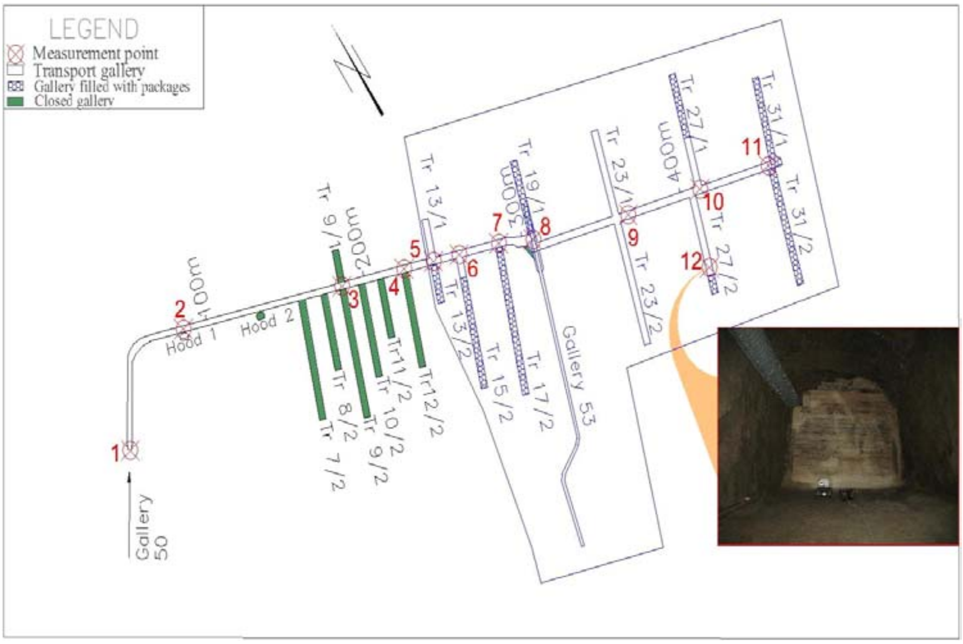

Fig. 1 – Location of the measurements points of the radon on the repository sketch.

2.1. MEASUREMENTS PERFORMED ALONG THE TRANSPORT GALLERY

This experimental session was the very first step and the most important one, having as objective to determine the natural transport pathways of radon inside mine galleries and to identify those points where radon accumulation reaches the highest concentration. The first session (09–10.07.2012) of measurements was done after 98 hours when the ventilation was turned-off. A set of three radon measurements was conducted, also environmental conditions (pressure, temperature and relative humidity) being monitored in parallel and recorded for the whole period of the experiments.

Determinations of radon concentration are shown in Figure 2. In point 1, positioned outside the repository, our device registered a radon concentration lower than the lowest detectable value (AMD) – 25 Bq/m^3.

There is an increase in the radon concentration as we move towards the center/end of transport gallery (points 2-6).

In point 8, located near the ventilation gallery (Gallery 53), there is a relatively large decrease in the concentration of radon due to natural circulation of air through the galleries (see Figure 2). Also, there is a decrease in the concentration of radon in points 7 and 9, located on both sides of the ventilation gallery (gallery no. 53).

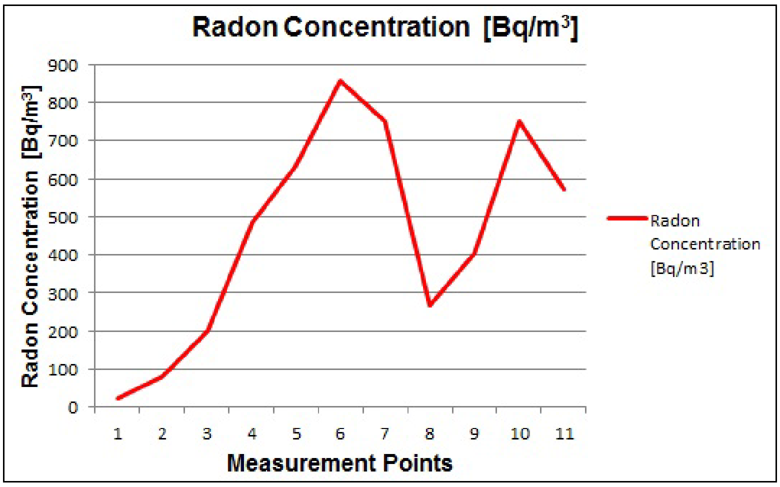

Fig. 2 – Variation of Radon concentration in transport gallery (gallery 50).

2.2. MEASUREMENTS PERFORMED IN FRONT OF THE GALLERIES 31/1 AND 31/2

During 09–11.07.2012, there were performed radon concentration measurements in the most remote point of gallery 50 (point 11), sited in front of the galleries 31/1 and 31/2 (data represented in the Figure 3).

The duration of each measurement was 6 minutes for phase 1–3, because we wanted to highlight, that for the same measuring interval of 6 minutes, the ventilation system is having an effect on the accumulation of Radon.

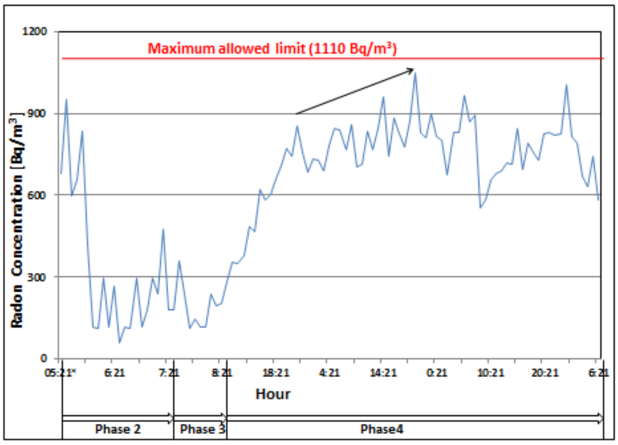

Fig. 3 – The evolution of Radon concentration (end point (11) of the transport gallery-50).

Measurements of radon concentration in this phase were conducted after a period of 4 days in which the ventilation system of the repository was stopped.
The average of 13 measurements of radon concentration (phase 1) is (678.2 ± 298.4) Bq/m^3, being below the maximum allowed limit for the operating personnel - 1110 Bq/m^3 regulated by the norm [11].

In phase 2, we can observe a rapid decrease of the radon concentration due to the influence of the ventilation system that was turned on. After turning off the ventilation of the repository, in the first 40 minutes (phase 3) we can observe no significant increase in concentration of the radon activity. Then, in phase 4, there is a relatively constant increase of radon activity concentration for 14 hours; the growth rate of the concentration is about 50 Bq/m^3/h.

Also, in phase 4, we can observe periods of growth and sudden decrease of radon activity concentration which can be explained by natural air convection that appeared in the gallery due to the tubes of the repository ventilation system which are located near the end of the measuring point.

Sudden drops are recorded at around 7 AM possible due to temperature variations outside the repository which can cause appearance of air currents in galleries 50 and 53, which can influence radon concentration measurement in selected points.

Radon concentrations recorded during the 71 hours of continuous measurements, after the 2 hours of ventilation, as an operational requirement, do not reach the maximum allowable limit of 1110 Bq/m^3 imposed by the legislation in force [11] for working spaces/controlled area in the nuclear field.

2.3. MEASUREMENTS PERFORMED IN THE END OF THE GALLERY 27/2

Taking into consideration the results obtained in session 2, between 11.09.2012–12.10.2012, there were conducted experimental measurements in three phases, in Gallery 27/2, at its end, considering that, in this point, the natural air currents formed in galleries 50 and 53 may have a less influence on the variation of radon activity concentration.

In phase 1.1 of the measurements, the scope was to observe the radon concentration before turning on the ventilation for 2 hours. In phase 1.2, in order to observe the decrease of radon concentration (ventilation turn-on), we set a measurement interval at 6 minutes. During this period, in order to increase the precision of the measurements we choose a 150 minutes measurement interval, as it is shown in Figure 4, phase 1. In phase 1.3, we set a measurement interval at 6 minutes and we observe the increase of radon concentration up to the maximum allowed limit (1110 Bq/m^3).

In phase 2, in the end of the gallery 27/2, for 25 working days and without ventilation, we registered 240 a measurement of 150 minutes each, for two major reasons:

– to put in evidence the evolution of the radon concentration in the end of the most distant gallery in operation from the entrance into the repository;
– to observe the evolution on a longer time period that allows us to collect of a set of comprehensive data on the pressure, temperature, humidity that might influence the radon concentration in time.

During the third phase, 9–12 October 2012, measurements were performed to observe the effect of ventilation which was turned on for 2 hours (keeping the same intervals like in the previous set of measurements and the data about the pressure, temperature and humidity).

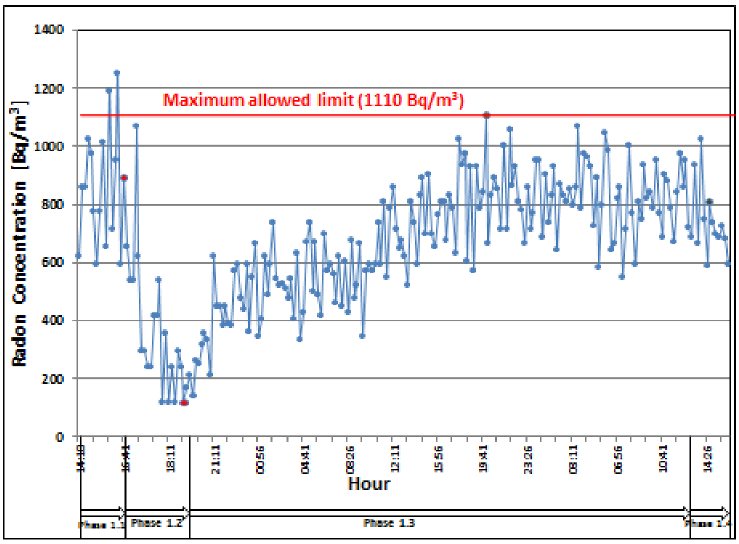

Fig. 4 – Phase 1 – Radon Concentration evolution in transversal gallery 27/2 during 11–14 September 2012.

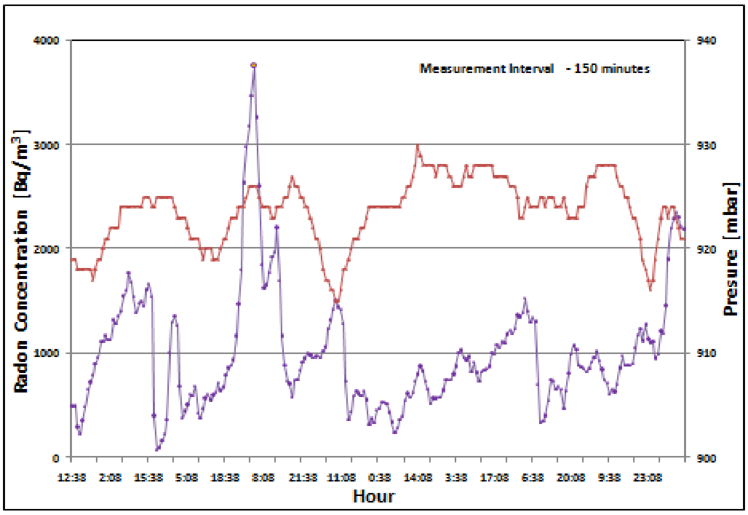

Fig. 5 – Phase 2 – Radon Concentration and presure evolution in transversal gallery 27/2 during 14 September–9 October 2012 (without ventilation).

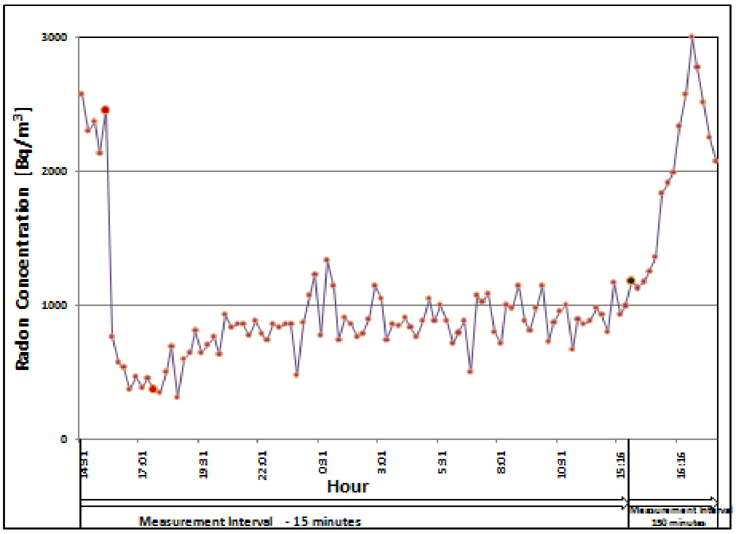

Fig. 6 – Phase 3 – Radon Concentration evolution in transversal gallery 27/2 between 9–12 October 2012.

3. DISCUSSIONS

The graphs in the present study, which represents the variation of radon concentration, recorded over approximately two months, show alternating periods of increasing and decreasing of the radon concentration. These variations were caused by the operation of ventilation, Fig. 3, Fig. 4 and Fig. 6 and by the presence of air currents inside the repository depending on the external weather conditions, currents that produce natural ventilation of the repository, Fig. 5.

It also can be identified periods in which the accumulation of radon reached a maximum concentration of 3752 Bq/m^3. This shows that in certain weather conditions (natural ventilation is negligible), the concentration accumulated in a transversal gallery exceed more than 3 times the maximum allowable operating personnel limit, imposing the existence and use of ventilation systems installed in the repository.

Considering the measurements taken during the 25 days, it can be seen a dependence of radon accumulation in gallery 27/2 on increased pressure, which can be explained by a dependence of the pressure values inside the repository on the weather conditions outside the repository. These conditions can be studied further on to see the influence of meteorological parameters outside the galleries of DNDR Baita-Bihor on the radon accumulation inside the transport gallery and inside the disposal galleries.

Monitoring results and onsite observations suggest that ionizing radiation hazards during disposal activities are relatively low; however, radon exposures needs intervention which can occur at some work locations.

Acknowledgments. We acknowledge support from the project PN II-PT-PCCA-2011-3.2-0334 - SARAWAD-BB.

The present study is also useful for the project EMERSYS activities (Toward an integrated, joint cross-border detection system and harmonized rapid responses procedures to chemical, biological, radiological, and nuclear emergencies), MIS-ETC code 774 (European Regional Development Funds, Romanian Ministry of Regional Development and Public Administration), being an useful input for radiological characterisation issues.

REFERENCES

1. Levinson, A.A., Bland, C.J., Dean, J.R., 1984. *Uranium series disequilibrium in young surficial uranium deposits in southern British Columbia*. Can. J. Earth Sci. 21 (5), 559–566.
2. European Communities (EC), 1990. *Commission Recommendation 90/143/EUROATOM of 21 February 1990 on the protection of the public against indoor exposure to radon.* Official Journal of the European Communities L-80, 26–28.
3. Charles Streeper, Julia Whitworth, J. Andrew Tompkins, *Lack of international consensus on the disposition and storage of disused sealed sources*, Progress in Nuclear Energy, 51 (2009) 258–267.
4. Cosma C., Szacsvai K., Dinu A., Ciorba D., Dicu T., Suciu L. *Preliminary integrated indoor radon measurements in Transilvana*, Isotopes in Environmental and Health Studies, 45:1-10, 2009.
5. Cosma C., Cucos-Dinu A., Papp B., Begy R., Sainz C., *Soil and building material as main sources of indoor radon in Baita-Stei radon prone area (Romania)*, Journal of Environmental Radioactivity, 116 , pp. 174–179, 2013.

6. Sainz C., Dinu A., Dicu T., Szacsvai K., Cosma C., Quindos L.S., *Comparative risk assessment of residential radon exposures in two radon-prone areas, Ştei (Romania) and Torrelodones (Spain),* Science of the Total Environment, 407 (15), pp. 4452–4460, 2009.
7. Rosabianca Trevisi, Claudia Orlando, Paolo Orlando, Mario Amici, Carla Simeoni, *Radon levels in underground workplaces e results of a nationwide survey in Italy*, Journal of Radiation measurements 47 (2012) 178–181.
8. Patitapaban Sahu, Devi Prasad Mishra, Durga Charan Panigrahi, Vivekanand Jha, R. Lokeswara Patnaik, *Radon emanation from low-grade uranium ore*, Journal of Environmental Radioactivity 126, 104–114, 2013.
9. M. Bochiolo, M. Verdoya, P. Chiozzi, V. Pasquale, *Radiometric surveying for the assessment of radiation dose and radon specific exhalation in underground environment*, Journal of Applied Geophysics 83, 100–106, 2012.
10. Hovav Zafrir, Susana M. Barbosa, Uri Malik, *Differentiation between the effect of temperature and pressure on radon within the subsurface geological media*, Journal of Radiation Measurements 49 (2013) 39–56.
11. Radiological Safety Norms in operational radiation protection mining and preparation of uranium and thorium, NMR 01 - Art 20. (1).