Carpathian Journal of Earth and Environmental Sciences, May 2013, Vol. 8, No. 2, p. 191 – 199

RADON MEASUREMENTS AND RADON REMEDIATION IN BĂIŢA-ŞTEI PRONE AREA

Constantin COSMA[1], Alexandra CUCOŞ DINU[1], Botond PAPP [1], Mircea MOLDOVAN[1], Robert BEGY[1], Tiberius DICU[1], Dan C. NIŢĂ[1], Bety D. BURGHELE[1], Dan FULEA[1], Ciprian CÎNDEA[1], Oana DUMITRU RUSU[1], Cristian MALOŞ[1], Liviu SUCIU[2], Gheorghe BANCIU [2] & Carlos SAINZ[1,3]

[1]Faculty of Environmental Science and Engineering, "Babeş-Bolyai" University, Fântânele street, no. 30, 400294, Cluj-Napoca, Romania, E-mail: constantin.cosma@ubbcluj.ro; alexandra.dinu@ubbcluj.ro; papp.botond@ubbcluj.ro; mircea.moldovan@ubbcluj.ro; robert.begy@ubbcluj.ro; tiberius.dicu@ubbcluj.ro; negru_frozen@yahoo.com; burghele.bety@ubbcluj.ro; fulea.dan@gmail.com; cipcin@yahoo.com; oana.rusu@ubbcluj.ro; cristian.malos@ubbcluj.ro; carlos.sainz@unican.es

[2]I.C.P.E. BISTRITA SA, Parcului street no. 7C, Bistriţa, Romania, E-mail: liviu@icpebn.ro; gheorghebanciu@yahoo.com

[3]Department of Medical Physics, Faculty of Medicine, University of Cantabria, c/Herrera Oria s/n., 39011, Santander, Spain, E-mail: carlos.sainz@unican.es

Abstract: Băiţa-Ştei was the largest uranium reserve in Romania with estimated reserves of 450,000 tons of high grade metal. It was a large open pit mine in the northwest of Romania (West Carpathian Mountains), situated at 123 km south-east of Oradea, the capital of Bihor County. The transport during the time of sediment by Crişul Băiţa water course increased the uranium and radium content in the river meadow. The building material from Crişul Băiţa river bed (stone, gravel, sand) was used as construction material for the houses. In addition, some people living on this valley and surroundings after the opening exploitation used as building material the uranium waste from this mine. Preliminary indoor radon measurement (grab samples) in the villages situated on the route of ore transport (Băiţa Plai - Ştei) shown high radon concentrations, until 5000 Bq m^{-3}. The new result obtained in this work in springer season 252 Bq m^{-3} is comparable with the annual means of 241 Bq m^{-3} and 229 Bq m^{-3} respectively, previously obtained, but more than twice times higher than the average value of 126 Bq m^{-3}, computed for Romania. About 3000 of etched CR-39 track detectors were used followed by a selection of 20 houses proposed for remediation where a systematic investigation regarding radon sources was performed. The measured indoor radon concentration in the surveyed buildings ranged from 40 to 4000 Bq m^{-3}. For experimental research, a representative pilot house was chosen. This house represents an example of a typical building from this area, with complex and various radon entry pathways which are correlated with the geology of soil. This building was chosen as pilot house due to the fact that it requires different ventilation systems or other remedial measures to be installed.

Keywords: indoor radon, CR-39 nuclear track detector, radon-prone area, radon diagnostic, remediation.

1. INTRODUCTION

Indoor Radon exposure of important population groups and associated health risks continue to be a major issue in EU Countries and in Radon Affected areas (Scivyer, 2007). It was stated that from the total annual radioactive dose received by the population over 40 % is due to inhalation and ingestion of radon and its decay products (UNSCEAR, 2006; ICRP, 2010; ICRP, 2012).

Research in radon field in buildings located in the uranium mines areas where the radon potential is higher are of great national and international interest. In many countries worldwide authorities targets monitoring all houses and implement effective methods of reducing radon where levels are elevated (Scivyer, 2007). In the field of the protection of houses against radon, several factors that could control the source and the behavior of radon in air inside building should be carefully studied. Radon concentrations in buildings widely vary depending on the ***underlying geological formations*** and ***characteristics of house***, like house structure, building materials, insulation or living habits.

The main source of indoor radon gas infiltration is from soil, rocks and type of building foundation. In addition, radon dissolved in water and radon emanating from building materials may contribute to increased levels of radon inside (Gunby et al., 1993; Cosma & Ristoiu, 1996).

Our previous researches on monitoring radon concentrations and potentially health risks effects in a series of buildings located in the extended region Ştei-Băiţa (Cosma et al., 2009; Sainz et al., 2009; Truţă-Popa et al., 2010; Cucoş (Dinu) et al., 2012; Cosma et al., 2013) reveals that the highest levels of radon were found in houses from Băiţa, Fînaţe, Cîmpani and Nucet villages.

The major aim was to identify both the most effective remedial measures that will be tested on pilot house and installed in 20 houses, as well as the factors that may affect their performance, including the geology and physical characteristics of the houses. The pilot house it is considered an example of a typical building from the area with complex and diverse radon entry pathways, in correlation with the geology of soil, requiring a number of ventilation systems or other remedial measures to be installed.

2.MATERIALS AND METHODS

2.1. Study Design

This study has been conducted in four localities **- Băiţa, Fînaţe, Cîmpani and Nucet-** located near the "Avram Iancu" and "Băiţa" uranium mines, in Bihor County, in North West part of Romania (Fig. 1), in the framework of an European project started in 2010 with aims to monitor radon concentration in a large number of buildings, in order to develop and implement various effective radon remedial methods to reduce indoor radon levels to as low as reasonably achievable. The main reasons why these four locations were chosen to extend our study are, firstly, because the preliminary results obtained showed the highest concentrations of radon in Romania, and secondly due to the influence of the local geology, this area being the closest settlements of uranium mines, from 2-5 km away. Moreover, these localities are situated on the riverbanks of Crişul Băiţa, which passes in the vicinity of the major operation point of uranium, now a non rehabilitated tailing from Băiţa Plai.

The indoor radon monitoring was conducted in two measurement campaigns performed during 2011 in 303 randomly selected houses. According to the project protocols, representative dwellings of the studied area were selected among those inhabited. Almost all the family houses monitored in the study are one-storey buildings. Radon measurement method consisted in passive detectors systematically distributed in ground floor rooms and cellars of each surveyed house, on average in 4 rooms / house.

Information about the characterization of the measurement site, such as type of building, year of construction, number of rooms, presence of cellar below the ground floor of the house, the use of uranium tailing as a construction materials, the distance from the uranium mines and living habits was routinely collected from the residents of each participant house.

2.2. Geological description of Băiţa-Ştei uranium mine area

Băiţa-Ştei uranium mine area has been carefully studied from the geological point of view. It is considered a metallogenic region situated in the North-Western part of the Bihor Mountains. Besides the exploration of the geological setting and discoveries of various ores, including uranium, many mining activities were promoted. This area contains a deep-seated Laramian pluton of granitic-granodioritic-dioritic composition and related dikes which penetrated various Paleozoic and Mesozoic sedimentary rocks (Stoici, 1983). They belong to several units consists of limestones (the Bihor unit), dolomites and limestones (the Codru unit), clays and sandstones (the Arieşeni unit). The rocks structure contains banatitic and batholith intrusives from Upper Cretaceous-Lower Paleogene with granites, granodiorites and pegmatites associated to a magmatic arc tectonic setting (Stumbea, 2003). The igneous rocks belong to the Hercynian and Laramian magmatic events. The Hercynian magmatism is represented by Permian rhyolites. Early volcanics are found in the Vermicular sandstones – feldspathic and oligomatic sandstones (Stoici, 1983). Therefore, the Băiţa village perimeter has a complex structural setting with various rock types from Carboniferous up to Neogene ages (Stoici, 1983).

The most significant uranium deposit was formed in the cupola of a granodiorite-granit pluton, installed in a tectonised, dominantly carbonatic, Mesozoic rocks – (Fig. 1). It is partially covered by the Arieseni Nappe with Permian detritic rocks (Stumbea, 2003). Băiţa Plai uranium deposit is situated in sedimentary permian rocks (sand stones and tuff) (Jude, 2006).

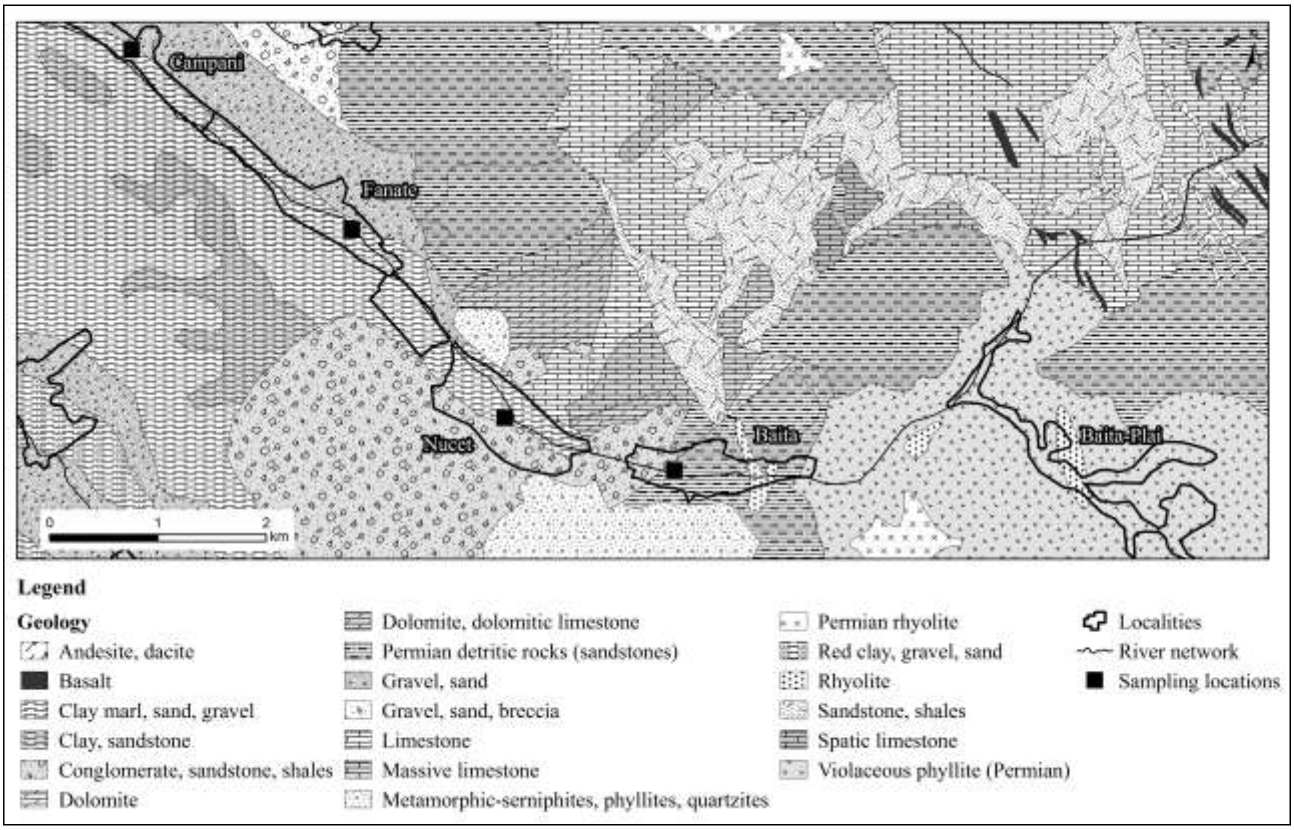

Figure 1**.** Geological map of Băiţa-Ştei uranium mine area and the localities selected for indoor radon measurement campaigns (simplified from Stoici, 1983).

2.3. Indoor radon measurement techniques

Radon measurements were performed by using solid state nuclear track detectors provided by Radosys Ltd. Hungary, based on CR-39 chips consisting of indoor passive monitoring between 1 month and 1 year. Each detector was personally deployed and collected by the members of the team within 303 houses investigated during this survey. The nuclear track detector consists of a cylindrical plastic vial provided with an appropriate lid. Onto the inside of the lid, clanged with adhesive can be found the 1 cm^2 CR-39 chip. The radon present indoor is able to enter the vial through the space created between the lid and the vial body. Thus inside, the alpha particles from radon decay hit the CR-39 chip living black tracks. Three months later, the usual exposure period, all detectors are returned to the lab, removed from the vial and etched in a 6.25 NaOH solution. During the next step, chips are 'read' using a Radosys microscope; according to the found track density the radon exposure and the indoor concentration can be calculated (Cosma et al., 2009; Sainz et al., 2009; (Cucoş (Dinu) et al., 2012).

The quality assurance of measurement method and the calibration of the detectors were periodically controlled at our Laboratory by using a small radon reference chambers (Cosma et al., 2009) as well as through the successful participation to several international intercomparisons (Janik et al., 2009; Jilek & Marusiakova, 2011; Cucoş (Dinu) et al., 2012).

2.4. Radon diagnostic in houses selected for remedial actions

This part of work presents the first investigations of this type at national level, being the first experience for Romania in radon diagnostics measurements and remedial measures in houses with high indoor radon concentration. In order to evaluate whether remedial measures is needed to reduce the radon levels and to determine the design of appropriate remediation method, detailed diagnostic measurements were carried out in 20 selected buildings.

Depending on indoor radon level, different radon mitigation strategies can be applied. Relatively cheap and not complicated measures, e.g. sealing of all visible cracks and leakages, reconstruction of old floors, are usually recommended as the first step, in the case of indoor radon concentrations not too high (not higher than 1000 Bq/m^3). More complicated remedial measures are recommended when the indoor radon concentrations are higher.

To design and install the most cost-effective radon control systems, a basic set of diagnostic procedures have been applied in our present research. At first, study of available documents and a visual inspection of the building with regard to relevant physical properties of individual elements of building structures have been done. This implies the history of the building, building materials and their origin, spatial arrangement, quality of different parts of the construction, mainly of the construction at the contact between the sub-soil and the building, tightness of windows and doors, inspection of visible cracks and leakages. Thereafter, it is necessary to accurately identify and quantify the main radon sources which contribute to indoor radon concentration (subsoil, building materials, water supply). Determination of radon potential of the building site is obligatory with the purpose to classify the building site area from the point of view of the risk of radon penetrating from the soil into the building (soil-gas radon concentration and soil permeability measurements). Hot spot air sampling from selected locations, suspected as a potential radon source (leakages, concrete slab cracks, wall-floor joints, technological penetrations through the building structure elements, etc) has been performed. Qualitative and quantitative analysis of radon entry pathways (simultaneous continual radon monitoring, radon blower door test) and analysis of indoor radon transport and distribution for different parts of the building have been done.

In order to develop, test and implement the most effective radon mitigation techniques, **a pilot house was selected from Fînaţe locality** (Fig. 2). This house represents an example of a typical building from the area with complex and diverse radon entry in correlation with the soil radon potential requiring a number of ventilation systems or other remedial measures to be installed.

Detailed radon diagnostic measurements were performed in and around this house during 2011.

Inspection of the house represents the first step of radon diagnostics. The family house is a ground floor house, which was built between 1976 and 1978. Building materials used for construction of the house was bricks, gravel, stones, sand and ballast from Criş-river, passes closed to uranium mine. The basement was filling with stones, from the former mining area (Băiţa-Plai) and local material (ballast from the river). The area of the building site is 3198 m². The cellar of the house was located below a bedroom. The whole house has been reconstructed in 2010. The floors are new, made from concrete covered by parquet or by paving. The windows are new and tight.

During radon investigations and remedial actions the measured part of the house was inhabited, the rooms were mostly closed. Only a part of the house was used for radon diagnosis investigations and remediation, consist from two bedrooms, one living, one bathroom and one cellar (Neznal & Neznal, 2011).

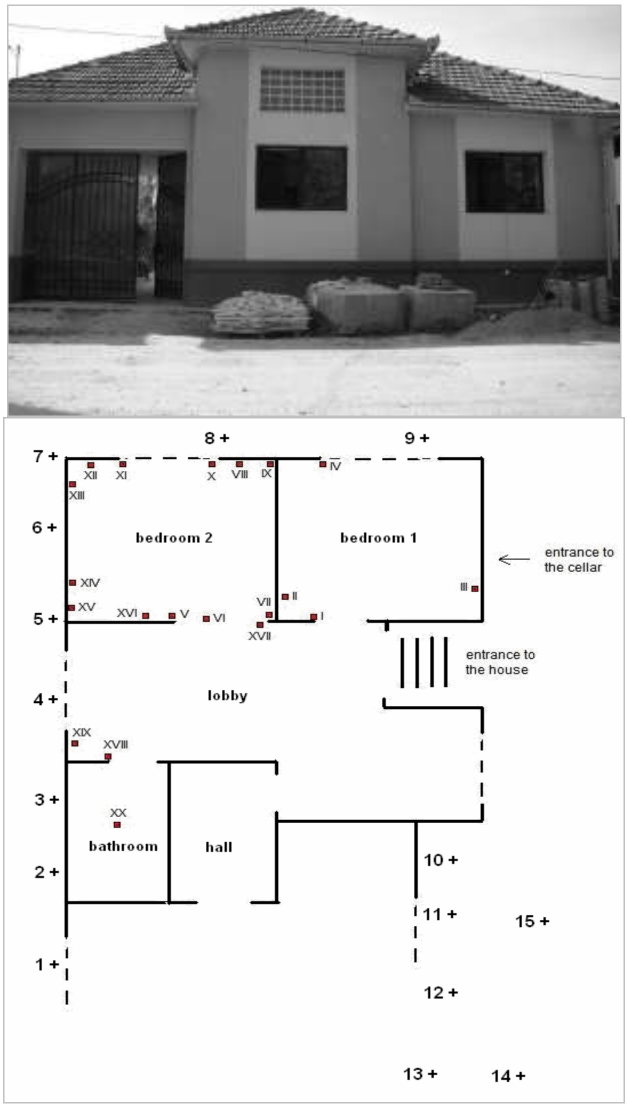

Figure 2**.** Pilot house (a). The disposition of the rooms in the studied part of the house, the locations of the radon in soil measurements and the locations of the leakages points in the contact of the subsoil and the building site (b).

Method of determination of the ***radon index at the building site;*** detailed description of the method is given in (Neznal et al., 2004; Barnet et al., 2008) in terms of radon potential (RP).

According to the model, the categories of radon index are the follow: if $RP < 10$, the radon index of the building site is low; if $10 \leq RP < 35$, the radon index of the building site is medium; and if $RP \geq 35$, the radon index of the building site is high (Neznal et al., 2004; Barnet et al., 2008).

Method of detection of the ***leakages in the contact between the subsoil and the building***. The method is composed on the collection of the air samples in places where leakages may appear (i.e. cracks, uptightness beside tubes going through the floors, etc.), and then measuring radon concentrations from samples using the same instrumentation as for the determination of radon concentrations from soil gas (Neznal et al., 2004; Barnet et al., 2008).

Continual measurements of indoor radon concentration in different parts (rooms) of the house enable to evaluate the most important radon sources and radon pathways in the building. This type of investigations can be performed by radon monitors as RADIM, Alpha Guard, RADON SCOUT, etc, which measure also climatic parameters

The main goal of ***gamma dose measurements*** in the house is to identify any inhomogenities of gamma radiation field and the detection of any gamma anomalies, which indicate the presence of radioactive materials in the walls, in the floors or in the filling. The investigations were performed by gamma dose rate meters (ex. Gamma Scout) that determine ambient dose rate in terms of [$\mu Sv \cdot h^{-1}$].

3. RESULTS AND DISCUSSION

3.1. Indoor radon results in the investigated area

A total number of 1128 passive detectors were collected during the period of 2010-2011. The distribution pattern of indoor radon concentrations depending on measurement campaigns in the 303 houses of Băiţa, Fînaţe, Cîmpani and Nucet localities is shown in the figure 3.

The distributions of log-transformed indoor radon concentration for the two measurments campaigns are normal, as confirmed by D'Agostino-Pearson test ($p = 0.5$ for the campaign I and $p = 0.4$ for campaign II, respectively). The differences between the results as function of the measurements campaign shows the seasonal influence (see Fig. 3).

Summary statistics of indoor radon measurements in dwellings on the ground floor of Băiţa uranium mine area (Băiţa-Nucet-Fînaţe, Cîmpani) are presented in table 1. Valid measurements were performed in 303 homes for 2 consecutive three months. For the **1st campaign** measurements were made during the winter session, while the measurements for **2nd campaign** were made in the spring season.

The average radon concentration (AM) depending on measurement campaigns for the entire investigated region were found to be bettween 351 and 252 Bq m^{-3}, respectively. The **2nd campaign** results are comparable with the means of 241 Bq m^{-3} and 229 Bq m^{-3} respectively, previously obtained by Sainz et al., (2009) and Cucoş (Dinu) et al., (2012), but more than twice times higher than the average value of 126 Bq m^{-3}, computed for Romania (Cosma et al., 2013). As it was expected, a significant statistically difference between log-transformed results of radon measurement according to the measurements campaign was observed, confirmed by Student's t-test for paired data ($p < 0.0001$).

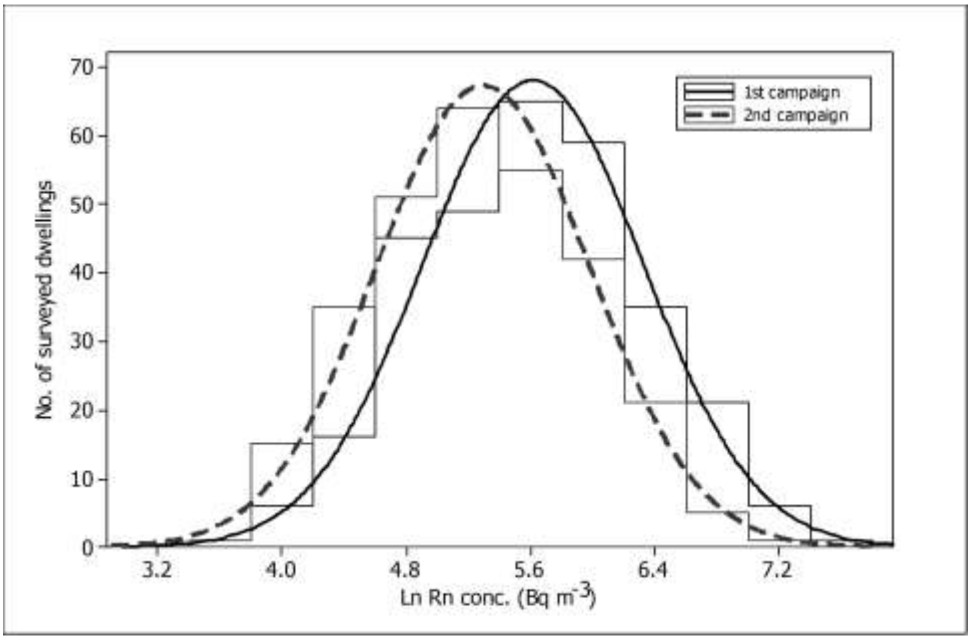

Figure 3. The normal distributions for the log-transformed indoor radon concentration in the two campaigns performed in Băiţa uranium mine area.

Table 1. Descriptive statistics of indoor radon concentrations in 303 monitored dwellings of Băiţa radon-prone area.

Indoor measurements campaign/ Period	**AM±SD [Bq m^{-3}]**	**GM±GSD [Bq m^{-3}]**	**Median [Bq m^{-3}]**	**Max. [Bq m^{-3}]**	**C.V. [%]**	**% > 300 [Bq m^{-3}]**
1st campaign (2010 Dec.-2011 Mar.)	351 ± 267	274 ± 2	273	1904	76	46
2nd campaign (2011 Mar.-2011 May)	252 ± 197	199 ± 2	195	1683	78	30

*** AM=Arithmetic Mean; SD=Standard Deviation; GM=Geometric Mean; GSD=Geometric Standard Deviation; CV=Coefficient of Variation.

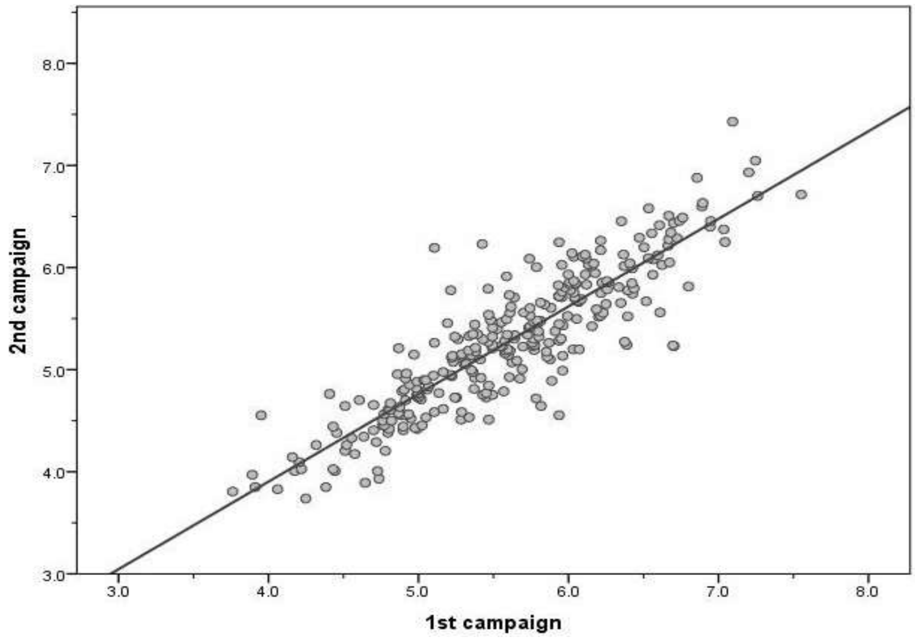

Figure 4. The correlation between the results for log-transformed indoor radon concentrations during the both measurements seasons.

For 46 % of all surveyed dwellings during the winter session the radon concentration exceeds the threshold of 300 Bq m^{-3}, as can be displayed in Table 1. A good correlation between the results for radon concentrations during the both seasons ($r = 0.87$, $p < 0.0001$) has been obtained, as shown in figure 4.

A detailed picture of the exposure to radon in Băiţa radon-prone area, the methodology of the survey, seasonal and regional variability, as well the influence of building characteristics of the indoor radon concentration in surveyed dwellings has already been described in more detail in Cucoş (Dinu) et al., (2012). Factors like the presence of cellar or the age of the building have been taken into account in order to perform analysis on the influence of these aspects in the entrance and accumulation of radon indoors.

3.2. Results for diagnostic in the pilot house

Results of direct in situ ***measurements of soil-gas radon concentration and of permeability of soil*** in the surroundings of the house are presented in table 2. From the evaluation of the results, $C_{Rn,75}$ = 40.9 kBq·m^{-3}, k_{75} = 9,9*10^{-12} m^2 and the value of the radon potential for the building site is RP = 40. This concludes to a high radon index (RI).

Table 2. Fanate 116A - Radon index of the building site.

Measuring point	**C_{Rn} [kBq·m^{-3}]**	**k (*10^{-11}) [m^2]**
1	0,6	1,4
2	8,9	< 0,00052
3	2,1	1,2
4	0,3	1,6
5	2,2	0,78
6	2,5	0,37
7	10,7	< 0,00052
8	**485,3**	**1,6**
9	32,6	< 0,00052
10	12,2	0,68
11	40,9	< 0,00052
12	96,5	0,99
13	46,2	0,27
14	12,5	< 0,00052
15	16,5	0,42

Results of the ***measurements to detect the leakages in the contact between the subsoil and the building*** are presented in terms of radon concentrations (C_{Rn}) in air samples collected in places and are summarized in table 3. Location of these points is shown in figure 2. Values which confirmed leakage points (i.e. > 1 kBq·m^{-3}) have been found mainly in the bedroom 2 and lobby, but also in the bedroom 1.

Table 3. Detection of leakages in the contact between the subsoil and the building.

Description (room) / no. of points	C_{Rn} range [kBq·m^{-3}]
bedroom 1 (4 points)	0.4 - **1.1**
bedroom 2 (12 points)	0.7 - **6.7**
lobby (3 points)	0.5 - **4.7**
bathroom (1 puncte)	0.5

Results of ***continual measurements of indoor radon concentrations*** in different parts (rooms) of the house are presented in figure 5, and a short statistical description of the continual measurement results (i.e. minima, maxima and mean values) are given in table 4.

Table 4. Statistical description of the continual measurement results (i.e. min., max. and mean values).

Room	Min. C_{Rn} [Bq·m^{-3}]	Max. C_{Rn} [Bq·m^{-3}]	Mean C_{Rn} [Bq·m^{-3}]
cellar	**25**	**2874**	**1131**
bedroom 2	**187**	**1425**	**889**
bedroom 1	**67**	**578**	**330**
lobby	**100**	**546**	**333**
bathroom	**56**	**731**	**395**

The highest radon concentrations have been found in the cellar, but the analysis of temporal changes indicates that there is no significant radon pathway of soil air from the cellar to the ground floor rooms. The temporal pattern of radon concentration is similar in all measured ground floor rooms and the indoor radon concentration in bedroom 2 is much higher than the indoor radon concentrations in other ground floor rooms. Therefore, the main pathway for the penetration of contaminated soil-gas into the indoor environment is therefore probably located in bedroom 2.

Results of ***external gamma dose rate measurements*** (detection of any gamma anomalies) are summarized in table 5.

Table 5. Gamma anomalies, results of dose rate (**D**) measurements.

Description (room)	D [µGy·h^{-1}]
bedroom 1: hot spot near the window, 0,5 m	**0.74**
outdoor wall near the entrance, basement	**0.28 - 0.40**

All observed anomalies are local. The background values in the surroundings of the house ranged from 0.1-0.2 µGy·h^{-1}. A little higher values were observed in the cellar, near the basement and on the floors of ground floor rooms (0.17-0.30 µGy·h^{-1}). Therefore, negative influence of contaminated building materials in the lowest part of the construction thus cannot be neglected.

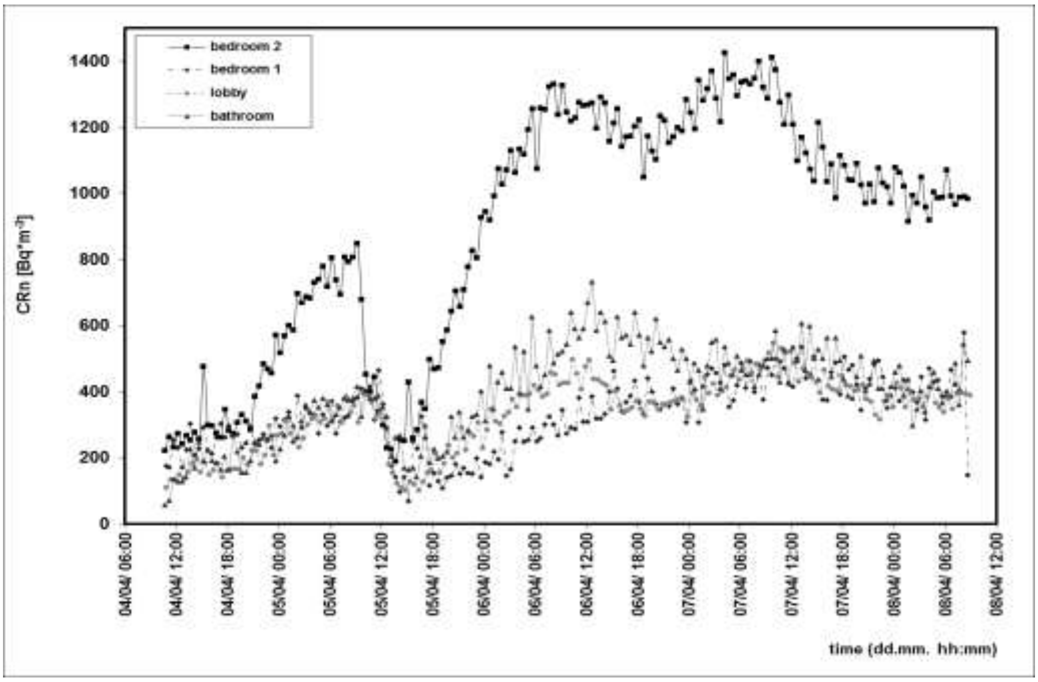

Figure 5. Continuous measurements of indoor radon concentration

3.3. Radon results in relationships with local geology

The high-radon levels inside the buildings from downstream uranium mine area can be caused by various reasons, the most significant being the mineralogical composition of soils under the houses, the building materials and the geological settings of the entire area. The relationship between geology and radon has been documented from the beginning of radon surveys and it is well known that the highest radon levels are mainly caused by the geological formations under the buildings. Geology controls the chemical composition of the rocks and soils from which radon is derived and, as pointed out by other authors (Quindos et al., 2004; Somlai et al., 2006), there is a linear dependency relation between the uranium content in soil and the radon concentration exhalation. Generally, as related by other researchers (Minda et al., 2009), the increased radon concentrations can be observed both on the indoor radon map and on the map of calculated geoparameters in the topographically confined areas corresponding to the extent of granitoid bodies but Băiţa Plai uranium mine is situated in sedimentary permian rocks where the main mineral containing uranium are pichblende and uraninite.

As mentioned above, different aspects of geology in the investigated Băiţa uranium mine area has been carefully studied and granitic-granodioritic-dioritic were found in the rocks structure. Therefore, from the analysis of indoor radon data it can be concluded that radon emanation from the ground is correlated of the lithology and tectonic structure of that area. However, further investigations are required on this topic and future work will be focused on more detailed studies.

Besides the influence of the geological formations, some parameters of the house structure also can influence the radon level. Radon exhalation from building materials has been studied since the early 70's as one of the important contributor to the indoor radon concentration. In the case of the houses from this area the population frequently used as building material the sand, gravel and stones from Crişul Băiţa river bed. This river passes through Băiţa Plai pit mine and the alluvial transport during the time enriched the uranium content of these materials from Crişul Băiţa Valley.

Based on our previously survey (Cucoş (Dinu) et al., 2012; Cosma et al., 2013), it can be stated that, in accordance with the investigations of other scientific researchers (Somlai et al., 2006; Minda et al., 2009), the radon concentration level in buildings in the vicinity of Băiţa uranium mine area appears to significantly exceed the both average values and recommended action levels (UNSCEAR, 2006; ICRP, 2010; ICRP, 2012).

4. CONCLUSIONS

About 3000 track detectors were used followed by a selection of 20 houses proposed for remediation where a systematic investigation regarding radon sources was performed. The measured indoor radon concentration in the surveyed buildings ranged from 40 to 4000 Bq m^{-3}

The distributions of log-transformed indoor radon concentration for the two measurments campaigns are normal, confirmed by D'Agostino-Pearson test and shows an seasonal influence.

The transport during the time of sediment by Crişul Băiţa water course increased the uranium and radium content in the river meadow. The building material from Crişul Băiţa river bed (stone, gravel, sand) was used as construction material for the houses. This fact combined with the high soil radon potential explains the elevated indoor radon levels.

Radon diagnostic measurements and investigations in pilot the house confirm that the most important source of indoor radon is the soil-gas from subsoil of the house (RP = 40). Two local gamma anomalies have been found in the basement and in the filling between the floor and the ground showing the presence of building material with increased content of uranium.

Acknowledgements. The survey was supported by the Sectoral Operational Programme “Increase of Economic Competitiveness” co-financed by The European Regional Development Fund under the project IRART 586-12487, Contract No. 160/15.06.2010.

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Received at: 11. 01. 2013
Revised at: 18. 04. 2013
Accepted forpublication at: 22. 04. 2013
Published online at: 26. 04. 2013