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INDOOR RADON MEASUREMENT IN FORMER URANIUM MINING REGIONS IN BULGARIA

Abstract:
Aim: Radon exposure situations have the characteristics of existing exposure situations since the source is unmodified concentrations of ubiquitous natural activity in the earth's crust. Human activities may create or modify pathways increasing indoor radon concentration compared to outdoor background. The mining and processing of uranium bearing minerals generate a variety of waste materials containing a number of radioactive and non-radioactive hazardous constituents. Conventional underground and open pit mining activities produce overburden, mineralized waste and barren waste rock, which are generally low in their uranium and thorium contents and are left at the mine site. The study focuses on regions of former uranium mining industries, where the radiological risk is higher.
Methods: The cumulative (passive) method was used for the study. The measurements are carried out by E PERM® system. Detectors are placed for approximately 6 months in randomly selected houses in former uranium mining sites - Sliven-villages area, Eleshnica and Bachkovo. These sites are situated in mountains in different part of Bulgaria.
Results: The results of indoor radon concentration for investigation villages range from 125 Bq/m3 to 4000 Bq/m3. The maximum concentration was measured in Bachkovo village in Rodopi Mountain. This village is the least affected by the former uranium mining industry.
Conclusion: The results prove assumption that former uranium mining sites are radon prone areas where radiological risk is higher. The level of indoor radon of residential buildings in areas with higher uranium availability is around or above the recommended reference radon levels. The maximum value of the concentration of radon is measured in areas less affected by uranium mining, proving the necessity to undertake a radon national survey to determine exact radon prone areas.
Key words: Radon concentration, long-term measurement, radon prone areas.

INTRODUCTION

Uranium mining in Bulgaria has 50 years history. For this period as a result of studies conducted throughout the country have discovered and developed 39 objects. (Fig.1) A numbers of exploration methods were applied: geological, geophysical, geochemical, technological and combined. The main ore deposits for underground extraction are: Buhovo near Sofia, Eleshnitsa, Senokos and Simitli in South-West Bulgaria, Vinishte and Smolyanovtsi in North-West Bulgaria, Sliven in Central Bulgaria, Smolyan, Dospat and Selishte in Rhodope Mountains.

Two hydro-metallurgical plants had been built in the town of Buhovo and in the village of Eleshnitsa for uranium ores processing and production of uranium concentrate (U_3O_8). The extraction and the processing of uranium ores in the Republic of Bulgaria were ceased based on secondary act of the Council of Ministers Decree (CMD) in 1992. Upon cessation of mining activities, hydro-ecological and radiological assessments and prognoses were prepared, as pre-project studies, work projects for the technical liquidation, the technical and biological remediation and for water purification and monitoring.

Figure 1. *Main uranium mining objects in Bulgaria*

In preparing the assessments are taken into account: the type of structural element, the number of construction,

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the area of the plots, the time of operation, proximity location, geographic and demographic characteristics of the region, the potential danger of pollution of air environment, soil, surface and groundwater reservoirs and drinking water sources and others. The objects are classified as regions with high, medium and low radiation risk based on assessments.

The uranium mining (uranium ore production and processing) activities resulted in the environmental contamination of the surroundings of the mining sites, both radioactive and non-radioactive. During the course of uranium ore processing the major part of the radioactive elements present in the ore (Th-230, Ra 226 and its decay products) is collected in the waste products resulting from the production processes. From these, the most dangerous radionuclide appears to be Ra-226 with 1617 years half-life, decaying down to Rn-222, a gas emitted into the atmosphere from the tailings ponds. The half-life of the latter is 3.8 days. Rn-222 decays to several short-lived products (Po-218, Pb-214, Bi-214, Po-214), to Pb-210 and Po-210, and finally to stable Pb-206. Radon is the key contamination agent of the air and in case of unfavorable climate conditions; lack of vertical circulation and redistribution in the surface-circulating air, its concentration significantly increases to values exceeding by far the permissible levels. Radon and the aerosols emitted into the atmosphere as well as the dust rising from the open, nonvegetated portions of the dumps stimulate the mechanical accumulation of radioactive dust in the adjacent agricultural and forest areas and the accumulation of long living and radiotoxic alpha- and beta-active nuclides such as Pb 210, Po-210 and Th-230 in values exceeding the permissible levels. This may cause enhanced external exposure and enhanced levels of.

The main source of indoor radon is Ra-226 in soil, outdoor radon concentration, building materials, and tap water. It is therefore assumed that the indoor radon concentration in uranium mining sites will be higher than other region in Bulgaria. Indoor radon may be significantly increased if residues form uranium waste rocks are recycled and use as construction materials. The study of indoor radon concentration focuses on regions of former uranium mining industries, where the radiological risk is higher.

MATERIALS AND METHOD

Design of the survey

The survey was carrying out in three villages near uranium mining objects in different regions of Bulgaria. The villages were chosen so that covers the different types of uranium mining object in Bulgaria with different sources of contamination. The survey was conducted in villages Eleshnitca, Sliven-villas area and Bachkovo (Fig. 1). The main purpose of this study was to demonstrate that the uranium mining sites are radon prone area of Bulgaria and assess associated annual effective dose of this group.

Eleshnitsa Village. The second milling plant “Zvezda” and two mines “Druzhba 1” and “Drizhba 2” are situated in Eleshnitsa. The village was the center of the southern mining region and is one of the sites with high radiation risk. Because the mine and milling plant were close to living areas and the village border, the radioactive and chemical pollution included ground and surface water, agricultural land, roads and living areas. The tailing pond of the Eleshnitsa milling plant Zvezda was established in 1969, and built in stages. All regular operation of the tailing pond was ended in 1995. The Zvezda milling plant and its tailing pond in Eleshnitsa were remediated under a European fund project (PHARE BG 9904-03-01-02). The project started in 2002 and ended in 2005. However some uranium-related activity is ongoing today, as the plant still extracts uranium from cation exchange resins that are installed throughout the country at old sites with water outflow and delivered in Eleshnitsa for processing into yellow cake. This is called the Facility for Regeneration Purification of Ion Exchange Resins and it is this activity that is at the origin of the small effluent pond seen to the north of the restored tailing pond. The population of Eleshnica is approximately 1420 inhabitants. The aim of the study was to gain an idea of the whole village. The village is conditionally separated in 5 regions. In the second and third region the houses are old and built with basements (before 1950). The houses in the fourth region are built on wetlands and have no basements. The houses in the first region are new.

Sliven-villas area. The mine “Sliven” is situated on the southern slope of the central part of Stara Planina, near the national park "The Blue Stones" in the suburbs of Sliven town. It has been discovered in 1955 using aero-gamma photography. It has been developed and operated since 1962 to 1981 using classical and later on combined technology with 2 vertical shafts and 14 adits (underground facilities). The South part of the field is included in the regulated recreation area of the town of Sliven. The technical liquidation had completed for two vertical shafts one adit in 1999 and eight more in 2002. After the uranium mining was ceased the expropriated forest and agricultural lands have been returned to their landholders. The biggest part of the land parcels have been building up, as the area is differentiated as so called resort area of the town of Sliven. After the ownership of the lands has been restored, the liquidated and filled – in with rock material form the waste rock heaps. Vertical shafts 1 now is situated in a private land ownership. The Villas area is divided into two regions – villas around shaft 1 and villas around shaft 2. Most homes in the Villas zone 1 are one-storey, old buildings, some of them are likely to be built with material from the mine waste piles. The houses in zone 2 are mainly two-storey and new construction. The region of survey is divided conditionally into three parts depending on the proximity of houses to the sites of uranium mining industry. The present study included houses near uranium facility to serve as a region with expected low concentrations of radon and to trace any contamination – zone 3.

Bachkovo Village. The village of Bachkovo is situated in the west part of Rhodopes Mountains, located amidst the old green forests of the mountain. In the area of Bachkovo are 2 mines adits, which located inside the mountain at about 2-3 km, where mining activities are conducted only 2-3 years. Practically no pollution from the mining activities in the village and this area is classified as an area with low radiation risk. The population of Bachkovo is 300 inhabitants. 1 km south of the village of Bachkovo is situated Bachkovo Monastery – the second in size and importance monastery in Bulgaria. Main objective was to cover the whole territory of the village. Detectors were placed in all parts of the village.

Measurement and evaluation

The cumulative measurements are carried out by E PERM® System . The E-PERM System consists of three components: (1) an electrostatically charged TeflonR disk called an electret which collects ions; (2) an ion chamber made of conductive plastic into which an electret can be loaded and (3) a reader to read the surface potential (voltage) of the electret. The method of the system E-PERM® is well designed, reliable, proven quality system, but is not suitable for large volume measurement because it is costly. LLT combination between camera and elecret are placed for long-term cumulative measurements (approximately 6 months) in randomly selected houses in different areas of the settlements in ground floor.

Table 1. *Data connected with survey*

Village	Period	Number of houses
Eleshnica	08.2008 to 01.2009	15
Sliven – Villas area	06.2009 to 03.2010	22
Bachkovo	07.2010 to 02.2011	20

Cameras are placed in rooms where the population resides most often. After the collection of the detectors, the compensation of voltage is read with reader. The electret voltage reader is a highly sophisticated electric-field sensor with a special receptacle into which the electret is placed. When the shutter is opened, the sensor can read the voltage on the electret surface without touching it. Readings of the electret voltage, before and after deployment provide an absolute number for unambiguous, quantitative determination of the ion collection accomplished by the electret due to radon in the chamber. Calculation is required to convert the two electret voltage readings (initial and final voltages) and the exposure period into a radon concentration value. First, the calibration factor (CF) must be determined using Equation (1):

$$CF = A + B\frac{(I+F)}{2},[\frac{V/d}{pCi/L}] \qquad (1),$$

where A and B are constants for a particular E-PERM configuration, and I and F are the initial and final electret voltages.

Equation (2) is used for calculating the radon concentration using the derived:

$$C = (\frac{(I-F)}{(CF*D)} - B*G)*k \qquad (2),$$

where CF is the calibration factor, C is the radon concentration and I and F are the initial and final electret voltages. D is the exposure period in units of days; k = 37 – correction factor for conversion of non-system unit pCi L^{-1} in the system unit Bq m^{-3}; B – gamma dose rate; G – correction factor - [(pCi/L)/(μR/h)].

To calculate annual effective doses from radon exposure, the formula recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) was applied:

Annual effective dose = AM (Bq m^{-3}) × 0.4 × 7000 (h) × 9 (nSv (Bq m^{-3} h)$^{-1}$), (3)

where AM is the arithmetic mean radon concentration in the units of Bq m^{-3}, the typical value of 0.4 was used as the equilibrium factor for radon indoors, a recommended value of 9 nSv (Bq m^{-3} h)$^{-1}$) was used to convert radon equilibrium equivalent concentration to population effective dose, and 7000 h or 80 % of home occupancy was assumed. The annual effective dose depends only on the AM.

RESULTS AND DISCUTION

The values of indoor radon concentration ranged from 200 to 1840 Bq m^{-3} in Eleshnica village. On the Fig. 2 is given the distribution of the measurement in Eleshnitca compared to the reference level (300 Bq m^{-3} - new European draft Directive). High values of radon concentration are measured in 66% of the total number of measurements.

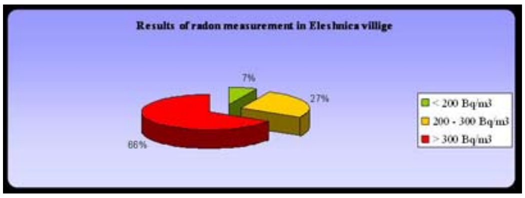

Fig. 2 *Indoor radon concentration measured in Eleshnica village*

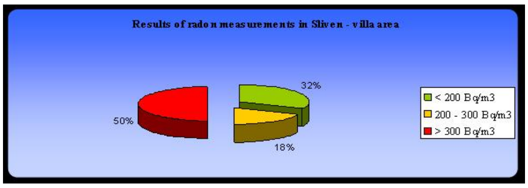

Fig. 3 *Indoor radon concentration measured in Sliven – Villas area*

The distribution of the measurement in Sliven – Villas area is given on the Fig. 3.

High values of radon concentration are measured in 50% of the total number of measurements Range of indoor radon concentration in thee part – Villas zone 1, 2, 3 is given in Table 2.

Table 2 *Range of indoor radon concentration in Sliven – Villas area*

Place	Range of indoor radon concentration [Bq. m^{-3}]
Zone 1	160 - 1160
Zone 2	95 - 1690
Zone 3	55 - 390

Concentration of radon in Villas zone 3 is lower than zone 1 and 2, which are close to mining facilities. In these areas, material from the waste rock heals is used for construction, possibly.

The values of indoor radon concentration ranged from 160 to 4560 Bq m^{-3} in Bachkovo village. The distribution of the measurement in Bachkovo is given 0n the Fig. 4. High values of radon concentration are measured in 58% of the total number of measurements.

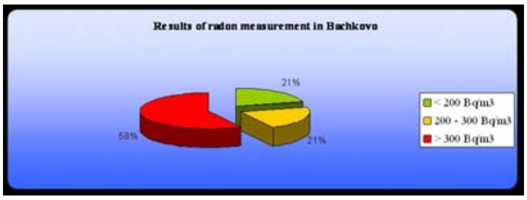

Fig. 4 *Indoor radon concentration measured in Bachkovo village*

The highest indoor radon concentration in measured in Bachkovo village. The annual effective dose from radon is evaluated using equation (3) and results are given in table 3.

Table 3 *Available information on radon exposure*

Village	Average radon concentration [Bq. m^{-3}]	Annual effective doses [mSv]
Eleshnica	675	17
Sliven – zone 1	430	11
Sliven – zone 2	590	15
Sliven – zone 3	150	4
Bachkovo	1010	25

In the surveyed areas, on average 50% of houses have higher concentrations of radon than the reference. The estimated annual doses are above the annual limit. This can be considered that uranium mining sites are radon prone area in Bulgaria.

CONCLUSION

The results prove assumption that former uranium mining sites are radon prone areas where radiological risk is higher. The level of indoor radon of residential buildings in areas with higher uranium availability is around or above the recommended reference radon levels. The maximum value of the concentration of radon measured in areas less affected by uranium mining, proving the necessity to undertake a radon national survey to determine exact radon prone areas.

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BIOGRAPHY

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UNUTRAŠNJA MERENJA KONCENTRACIJE RADONA U REGIONIMA BIVŠE EKSPLOATACIJE URANIJUMA U BUGARSKOJ

Kremena Ivanova, Victor Badulin

Rezime:
Cilj istraživanja: Izlaganje radonu ima slične karakteristike kao izlaganje uobičajenim koncentracijama koje su posledica opšte prisutnih prirodnih aktivnosti u zemljinoj kori. Antropogene aktivnosti mogu da prouzrokuju promene koje povećavaju unutrašnju koncentraciju radona, u poređenju sa spoljašnjim, prirodnim fonom. Iskop i obrada minerala za ekstrakciju urana generišu raznolike otpadne materijale koji sadrže brojne radioaktivne i neradioaktivne komponente. Aktivnosti vezane za konvencionalne podzemne i površinske kopove prouzrokuju nastanak jalovine, mineralizovanog i inertnog otpada koji se ostavlja na mestu nastana, a sadrži relativno niske koncentracije uranijuma i torijuma. Ova studija se odnosi na regione u kojima je nekada postojala eksploatacija urana i u kojima je povećan radiološki rizik.
Metodi: U ovoj studiji upotrebljen je kumulativni (pasivni) metod. Merenja su izvedena primenom E PERM® sistema. Detektori su postavljeni u trajanju od približno 6 meseci u slučajno odabranim kućama koje se nalaze u oblastima nekadašnje eksploatacije uranijuma: Sliven – seosko područje, Elešnica i Bačkovo. Ovi lokaliteti se nalaze u planinskim oblastima različitih delova Bugarske.
Rezultati: Vrednosti unutrašnje koncentracije radona u ispitivanim selima iznosila je od 125 Bq/m3 do 4000 Bq/m3. Maksimalna vrednost izmerena je u selu Bačkovo koje se nalazi na planini Rodopi. Ovo selo je bilo najmanje izloženo uticaju nekadašnje eksploatacije urana.
Zaključak: Rezultati nameću zaključak da se lokaliteti nekadašnjih rudnika uranijuma oblasti sa povećanim prisustvom urana u kojima je radiološki rizik izraženiji. Nivo unutrašnjih koncentracija urana u stambenim objektima oblasti bogatih uranom je oko ili iznad preporučenih, referentnih nivoa koncentracija urana. Maksimalne vrednosti koncentracije urana su izmerene u oblastima koje su bile manje izložene aktivnostima njegove eksploatacije, što potvrđuje potrebu da se preduzme istraživanje oblasti sa povećanim prisustvom urana na nacionalnom nivou.

Ključne reči: Koncentracija radona, merenja dugoročne ekspozicije, izložene oblasti.