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Radon Hazards in Relation to Elemental and Isotope Composition of the Geological Structures in the Lubelskie Voivodeship

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ABSTRACT

The study involved investigation of the relationship between the radon concentrations in the ground air – and thus in the indoor air – and the geological structure of the Lubelskie Voivodeship (eastern Poland). Both passive and active methods were used for measuring the radon concentrations in coal, phosphate and chalk mines, caves, wells as well as indoor environments. The study also included elemental, uranium and lead isotope analyses of rocks. The performed research showed that Paleogene and Mesozoic sedimentary rocks rich in radionuclides are the sources of radon in the Lubelskie Voivodeship. In the case of the buildings located in proximity to such rocks, characterized by relatively high radon exhalations, radon remediation methods are recommended. Already at the designing stage of buildings, the measures which protect against the hazardous radon gas should be applied.

Keywords: rocks, radon sources, emanations, radon exhalations, indoor environment

INTRODUCTION

Ionizing radiation is an inherent component of the environment. However, the high background level of this radiation can become dangerous to health and thus can significantly degrade the indoor air quality. The main contributor to the indoor radiation exposure is radon, a natural radioactive gas. Gaseous radon and its solid decay products very often account for half the total dose from all radiation sources [UNSCEAR 2008]. Radon comes from alpha decay of radium and is an intermediate step in the uranium radioactive decay chain. The indoor radon concentrations mainly depend on the geological composition and structure of the ground under the buildings as well as the applied construction materials. In the case of the building ground, the radon entry to the indoor air environment is dependent on the most convenient paths of radon migration, which are determined by the tectonics and hydro-geological conditions [Przylibski et al. 2011, Gazda et al. 2016].

The paper presents the first attempt to correlate the geological structure of the Lublin region (eastern Poland) with the radon exposure in buildings and in other sites like mines, caves, wells. Local tectonic, lithological and hydrogeochemical conditions of the ground and the resultant distribution of radon sources were taken into consideration.

METHODS

The measurements of radon concentrations were performed in coal, phosphate and chalk mines in the Lubelskie Voivodeship, Poland. The geological and geochemical structure of the analyzed sites differed, thus influencing the radon transport. The passive method with the use of solid state nuclear track detectors with CR-39 foil (Radosys, Hungary) was employed for the measurements of radon concentrations. The exposure time of detectors ranged from 1 to 1.5 month, depending on the outcome of the measurements conducted with the active method using AlphaGUARD (Genitron, Germany) professional radon monitor (see Figure 1).

After exposure, CR-39 detectors were chemically etched and alpha particle tracks were counted using automatic microscope system (RADOSYS Radometer 2000, Hungary). On the basis of the obtained track density, the average radon concentration during exposure was determined.

Following the grinding and digestion of rocks, the chemical analysis of elements was performed. The digestion of ground rocks was performed in HF digestion vessels with the Microwave 3000 (Anton Paar, Austria) microwave extractor/digester. The digestion involved the use of a mixture of nitric and hydrochloric acid Suprapur grade (1:3, v/v, Merck, Germany). JobinYvon JY-238 ICP-OES system calibrated with CertiPUR-VIII (Merck) standard solution was employed for the final analysis of elements. Perkin Elmer NexION 300D ICP-MS system was used for analyzing the lead and uranium isotopes

RESULTS

Table 1 presents the obtained concentrations of radon in the underground air at the representative measurement sites in the Lubelskie Voivodeship. According to the date in Table 1, in the excavation cave I containing gaize rocks and bedrocks, the radon concentrations ranged from 920 to 1930 Bq/m^3. The highest radon concentrations were measured in the air of an abandoned phosphate mine (~5500 Bq/m^3) where phosphorite rocks and sandstone were dominant. On the other hand, lower values of radon concentrations were found in the excavation cave II (~3800 Bq/m^3) and chalk mine (~5000 Bq/m^3), where gaize rocks and chalkstone occurred, respectively. The air of an active, ventilated coal mine, in which sandstones and claystones constituted the waste rocks, was characterized by the lowest radon concentrations (~100 Bq/m^3).

Table 2 presents the contents of selected elements in phosphorite rocks originating from the phosphate mine and in gaize rocks taken from the excavation cave I. As can be seen from Table 2, the contents of lead and bismuth in sedimentary rocks from excavation cave I are much higher than in the case of phosphorite rocks from the phosphate mine. In relation to the excavation cave I, the content of strontium and nickel in phosphorite rocks is much greater. On the other hand, cobalt and cadmium are at a similar level in the analyzed locations.

Table 3 presents the uranium and lead isotope contents in phosphorite rocks from phosphate mine and gaize rocks from excavation cave I.

Table 3 shows that much higher content of uranium isotope ^{238}U was found in the sedimentary rocks from excavation cave I than in phosphorite rocks from the phosphate mine. The contents of lead isotopes ^{208}Pb and ^{210}Pb were found to be also higher in the sedimentary rocks from excavation cave I, in relation to phosphorite rocks. These rocks exhibit a similar content of uranium isotope ^{235}U (< 10 ppm). The rocks from excavation cave I have a greater content of lead isotope ^{208}Pb, con-

Figure 1. Measurements of radon concentrations carried in one of the caves using AlphaGUARD and passive CR-39 detector

Table 1. Measurements of radon concentrations

Measurement site	Stratification	Rocks	^{222}Rn [Bq/m^3]
Coal mine	Upper Carboniferrows (Westphalian)	Coal Sandstone Claystone	80–120 ± 7–10
Phosphate mine	Middle Cretaceous	Sandstone Phosphorite	5499–5709 ± 437–447
Chalk mine	Upper Cretaceous (Maastrichtian)	Chalkstone	4990–5150 ± 190–224
Excavation cave I	Upper Cretaceous (Paleocene)	Gaize Bedrock	920–1930 ± 40–80
Excavation cave II	Paleocene	Gaize	3780–3820 ± 120–170

Table 2. The contents of selected elements in ground rocks

Site	Bi [ppm]	Cd [ppm]	Co [ppm]	Ni [ppm]	Pb [ppm]	Sr [ppm]
Phosphate mine	<0.5 ±0.2	<0.2 ±0.2	1.2–1.5 ±0.1	119–132 ±0.5	<0.5 ±0.1	304–357 ±0.5
Excavation cave I	0.81–1.27 ±0.2	<0.2 ±0.2	0.8–1.7 ±0.1	17.5–23.8 ±0.5	3.1–4.6 ±0.2	246–248 ±0.5

Table 3. Uranium and lead isotope contents in ground rocks

Site	^{238}U [ppb]	^{235}U [ppb]	^{208}Pb [ppb]	^{210}Pb [ppb]
Phosphate mine	<10–25 ± 10	<10 ± 5	0.74–0.98 ± 0.05	0.35–0.44 ± 0.05
Excavation cave I	95–134 ± 10	<10 ± 5	6.4–8.3 ± 0.05	2.9–3.8 ± 0.05

stituting the decay product of ^{232}Th, than in phosphate mine; indirectly, this shows a higher content of ^{232}Th in the rocks from excavation cave I. Presumably, higher thoron (^{220}Rn) concentration can be found in excavation cave I, but measurements have to be carried out in order to confirm this estimation.

The content of lead isotope ^{210}Pb, being a direct decay product of ^{210}Bi, corresponds o the total bismuth content in rocks. The sedimentary rocks from excavation cave I exhibit higher uranium, bismuth and lead contents than phosphorite rocks. This may indicate that more intensive volcanic processes occurred in the Paleogene when phosphorites were formed than in the Albian/Cenomanian period. The concentrations of radionuclides are influenced by the age of sedimentary rocks and phosphorites to a lesser extent.

DISSCUSION

The performed study shows that Paleogene sedimentary rocks rich in radionuclides, which form the Lublin graben and Mesozoic sedimentary rocks of Lublin Basin constitute the source of radon in the Lubelskie Voivodeship.

The radon concentrations in schools and other living premises were analyzed out in the Lubelskie Voivodeship over the period of 2009–2016. The obtained results showed that the value of 300 Bq/m^3 recommended by EU Directive 2013/59/Euroatom was exceeded in several cases [Kozak et al. 2012, EU Directive 2013, Műllerová et al. 2016]. It should be added that the living premises and classrooms were naturally ventilated and used for different purposes. They were located on different stories, varied in volume, and in respect to the materials used for the construction of walls and floors.

Simultaneously, the research on the radon concentrations found in wells, underground mine excavations and groundwater intakes indicated values ranging from 100 to 6000 Bq/m^3, whereas the highest measured concentrations reached over 14000 Bq/m^3. Using the obtained data, the background concentrations of radon occurring in underground facilities of the sub-surface of the Lubelskie Voivodeship lithosphere were determined lithosphere [Gazda et al. 2012, Gazda et al. 2016]. The radon concentrations up to 300 Bq/m^3, resulted from the geochemistry and lithology of the considered area (content of radionuclides). Abnormally high radon concentrations may result from the zonation of tectonics and the hydrogeological conditions [Kozak et al. 2005, Gazda et al. 2016]. The anomalous values of radon concentrations are similar to the ones noted in the Sudetes [Przylibski et al. 2011, Kozak et al. 2011], containing igneous and metamorphic rocks (Figure 2). Crystalline rocks in the Lubelskie Voivodeship are usually found at the depth of about 8 km in

Figure 2. The Lubelskie Voivodeship and its vicinity (A) and Sudetes (B)

the eastern part (Nałęczów, Annopol) up to 1 km eastward, in the vicinity of Chełm [Żelichowski 1984]. Radon emissions could not stem from the carboniferous sediments. The measurements conducted in the excavations of Bogdanka hard coal mine (Table 1) indicated the concentrations at the background level of lithosphere characterizing the Lubelskie Voivodeship. This observation is also confirmed by a systematic monitoring carried out by the mine management as well as low content of uranium and thorium found in carboniferous rocks (mudstones, claystones, sandstones, coal) (Gazda 2005). Above the Carboniferous rocks, within the Lublin Basin, there are carbonate, carbonate-silicate, and carbonate-clay Jurassic, Cretaceous and Palaeogenic sediments as well as thin layers of Neogene, Pleistocene and Holocene sediments. The only components of these rocks which could potentially exhibit elevated content of radionuclides in the areas under study include phosphorites, which significantly accumulated in the Cretaceous layers and – to a lesser extent – in the intermediate between Cretaceous and Paleogene. The participation of radiogenic potassium in this potential, contained in glauconite, cannot be ruled out. The potassium content in glauconite reaches up to 8% (Krzowski, 1993). This mineral forms associations with phosphorites in Albian, Cenomanian, and Peleogene. The share of radiogenic potassium radiation dose in glauconite will be considered in a separate paper. This issue will be important for the region due to the possibility of initiating surface exploitation of Eocenic glauconite sediments with amber and phosphorites in the near future. The measured radon concentrations in an abandoned phosphorite mine (Table 1) were the highest (5499–5709 Bq/m^3) out of all monitored mining excavations. The conducted investigation, related to the uranium content in the Cretaceous phosphorites (Table 2), indicates lower levels than in the case of Paleogene phosphorites, but the observed radon concentration level is also connected with the amount of these phosphorites contained in the rock mass. High concentrations of radon characterizing the Cretaceous excavations (4990–5150 Bq/m^3) – which are free of phosphorites – result from lower, relatively shallow (150–200 m) layers of Albian/Cenomanian with phosphorites. The main source of radon in the Lubelskie Voivodeship can thus be attributed to the emanations from the lower Upper Cretaceous level (Albian/Cenomanian) and to a lesser degree, the upper Cretaceous-Paleogene strata.

The conducted studies indicate the source of radon emanations; their impact on the internal environment depends on the privileged circulation routes which are determined by tectonics and hydrogeological conditions [Gazda et al. 2016].

CONCLUSIONS

The indicated relatively high local radon concentrations in the subsurface of the Lubelskie Voivodeship originate from the presence of Cretaceous-Paleogene sedimentary rocks, rich in radionuclides. A more detailed analysis of indoor environments of the existing and designed structures is necessary. This is vital in regard to the currently developed passive houses, recuperation, closed ventilation and air conditioning systems, ground-coupled heat exchangers, etc. The designing stage of such solutions should be preceded with a geological research as well as measurements of radon concentrations in the ground air, especially radon index RI (as a function of radon concentration in soil gas and soil permeability) should be determined at the areas where new buildings are planned. These guidelines are related to every area characterized by excessive radon concentrations in the ground air, not only to the considered locations in the Lubelskie Voivodeship.

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