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Distribution of radon activity in the atmosphere above Wzgórza Niemczańsko-Strzelińskie (South-West Poland) and its dependence on uranium and thorium content in the underlying rock and indirect ground basement

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Abstract
Radon activity in the atmosphere and its behavior in the environment have been investigated using LR-115 nuclear track detector. The complex geological structure of Wzgórza Niemczańsko-Strzelińskie (south-west Poland) enabled this problem to be studied in various geological conditions. The eU and eTh content in rocks and soil was measured by gamma-spectrometer GR-320. Uranium content of bedrock reached its maximum value of 15 ppm in the case of quartz-graphite schist. Thorium reached its maximum value of 35 ppm in the case of granodiorite. Radon activity was measured by means of long-term exposure of LR-115. The mean value of atmospheric radon activity was 21 Bqm^{-3} in the air 2 m above the ground surface. The highest radon activities were measured in the area of granite and quartz-graphite schist outcrops and in the area of mylonitic rocks of the Niemcza Zone. Radon activity in close to ground cup detectors varies from 25 to 300 Bqm^{-3}, these values depend on uranium and thorium content in indirect ground basement (soil and weathered rocks). Not only uranium and thorium content but also rock disintegration due to tectonic events (shear zones) influenced atmospheric radon activity. Seasonal variation is not strong, although higher values were measured in the autumn-winter period.

Key words *radon – atmosphere – uranium – thorium – rock*

1. Introduction

Radon is released into the atmosphere mainly from soils and underlying rocks, from ground waters (especially thermal ground waters), but also from oceans, natural gases, caves and mines (Gesell, 1983; Khatir Sam and Holm, 1995; Lozano *et al.*, 2000; Jha *et al.*, 2001; Papastefanou, 2001). Spatial variation of outdoor radon activity and its dependence on the geological conditions, on the type of cover of Earth surface (ocean, ice cap or snow cover) and on the variations in soil moisture (Grasty, 1991) was investigated in many areas worldwide (Gesell, 1983; Gundersen, 1991). There is a large number of measurements of radon activity in the air above the ground surface, but due to the application of different methods, the results are not compara-

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ble. Gesell (1983) compared the data from the area of United States, where the average radon activity in the air varies between 4-15 Bqm^{-3}. In Canada outdoor radon activity, measured by one of the passive methods with an exposure period of 3 months, varied from 11 Bqm^{-3} to 67 Bqm^{-3} (the mean value for Manitoba is 59 Bqm^{-3} and Saskatchewan – 61 Bqm^{-3}) (Grasty, 1991). According to UNSCEAR (1983 fide Wilkening 1990) the typical radon activity in the air above the ground surface averages 10 Bqm^{-3}.

The aim of this investigation was to enrich the knowledge of radon behavior and to construct a map of spatial variation of radon activity on the eastern area of Foresudetic Block.

2. Geological setting

Wzgórza Niemczańsko-Strzelińskie are located in the eastern part of the Foresudetic bloc (SW part of Poland). This area includes the periphery of the Sowie Góry Block, Niemcza Zone, metamorphic of Niemcza-Kamieniec Ząbkowicki, which is extended on the NE from the Niemcza Zone, and a crystalline massif of Wzgórza Strzelińskie (fig. 1). The North-East part of Sowie Góry Block is composed mainly of paragneisses and migmatic gneisses. In the gneisses small bodies of amphibolite, gabbro and serpentinite are inserted (Dziedzicowa, 1987). In the vicinity of the boundary of Niemcza Zone there are intrusions of quartz monzodiorite, which belongs to the plutonic rocks of Niemcza Zone (Dziedzicowa, 1987).

Niemcza zone is extended along the east edge of Góry Sowie Block. It is the zone of dislocation of the mass of rocks and is built of mylonites originated from deformations of gneisses of Góry Sowie (Scheumann, 1937 fide Dziedzicowa, 1987; Mazur *et al.*, 1995). The mylonitic rocks include the small bodies of amphibolites, quartz-graphite schist, the enclaves of gneisses and intrusions of plutonic rocks (Mazur *et al.*, 1995). In the south part of Niemcza Zone there are vast outcrops of serpentinite and gabbro (Puziewicz and Radkowska, 1990).

The eastern edge of Niemcza Zone adjoins the series of mica schist or more general series of metamorphic rocks. This area is known as a metamorphic of Niemcza-Kamieniec Ząbkowicki and its contact with the crystalline massif of Wzgórza Strzelińskie (Strzelińskie Hills) in the eastern part is covered with Quaternary sediments (Dziedzicowa, 1966). Metamorphic of Niemcza-Kamieniec Ząbkowicki is built mainly of mica schist with the inserts of quartz-feldspathic schist, quartz-graphite schist, amphibolites and marbles (Dziedzicowa, 1966).

Between the metamorphic of Niemcza-Kamieniec Ząbkowicki and Wzgórza Strzelińskie, near Górka Sobocka village, there are outcrops of granite intrusion and its southern metamorphic cover – gneisses of Wzgórza Lipowe (Lipowe Hills) (Wojnar, 1977; Bartz and Puziewicz, 1999). According to Oberc-Dziedzic and Szczepanski (1995) the granite from Górka Sobocka is the westernmost part of the crystalline massif of Wzgórza Strzelińskie.

The series of crystalline rocks of the Wzgórza Strzelińskie massif can be divided into four groups (Oberc-Dziedzic, 1991): gneisses, the elder schist series, the younger schist series and plutonic rocks. The most common are gneisses. The rocks of the elder schist series (amphibolites, mica schist, limestone and marbles) appear in gneisses and are crimped with the rocks of younger series. The younger schist series, named Jeglowa series, is composed of quartzite, quartz-sericite schist, mica-sillimanite-quartz schist. The youngest group of rocks are plutonic rocks: quartz diorite, tonalitie, granodiorite and granite. Granite predominates in the northen part of massif (Oberc-Dziedzic, 1991). On the whole area of investigation there are the intrusions of Tertiary basalts.

Complex geological structure of this area, a wide variety of types of rocks and the presence of tectonic zone between Sowie Góry Block and crystalline massif of Wzgórza Strzelińskie (Mazur *et al.*, 1995), enabled the discussed problem to be studied in various geological conditions.

3. Methods

The fundamental equipment of this investigation was the LR-115 detector (production of Kodak), which is one of many types of solid

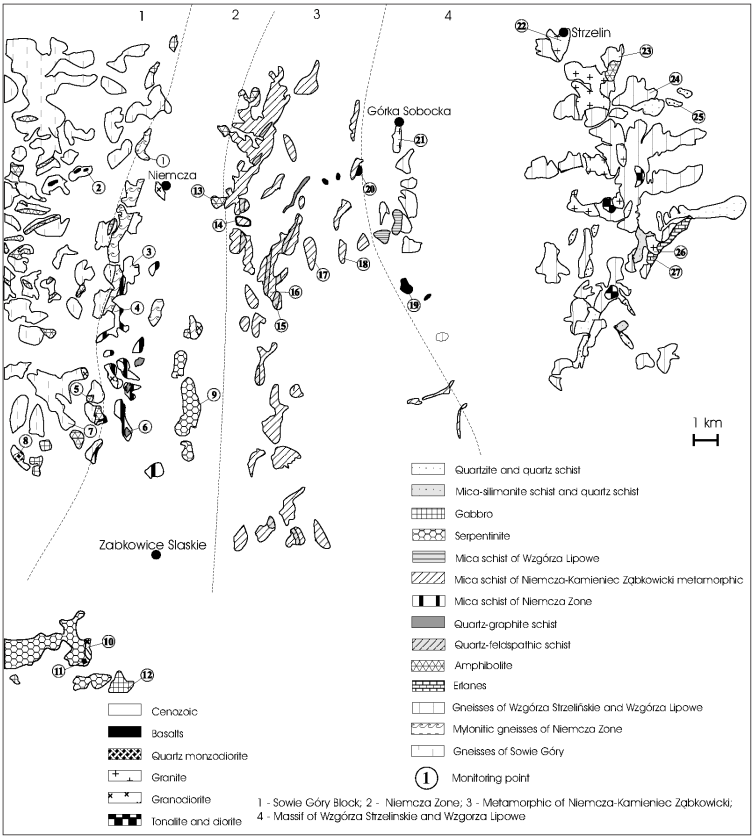

Fig. 1. Geological scheme of Wzgórza Niemczańsko-Strzelińskie (scheme prepared on the basis of the geological maps made by Badura, Berezowska, Cwojdzinski, Dziemianczuk, Gawronski, Gazdzik, Jerzmanski, Oberc, Trepka, Walczak-Augustyniak, Wòjcik).

state nuclear track detectors. The application of solid state nuclear track detectors is based on the creation of structural defects on the sensitive surface of the detector due to alpha-particle hit against it (Fleischer *et al.*, 1965). The structural defects were enlarged during the process of etching, which enabled us to observe them even at slight magnification.

A LR-115 detector was used for measuring the radon activity in the air (both in the open air and inside of a closed can). According to the information of its manufacturer, the LR-115 detector records the alpha-particles in a range of 1.2-4.8 MeV, meaning that the alpha-particles emitted in a well-defined distance from the detector could be registered. This characteristic facilitates the elimination of recording alpha-particles emitted from the solid products of radon decay.

In order to carry out the measurements of *radon activity in the outdoor air*, the detectors were fixed to the inner surface of a black plastic cup (of 8 cm diameter). The plastic cups provided shelter from the sun light and precipitations. We chose 27 monitoring points were situated on the outcrops of the different types of rocks. Each monitoring point consisted of 4 cups fixed 2 m, 1 m, 0.5 m and 0.05 m above the ground surface (fig. 2a,b). The higher the level above the ground surface where the detector was fixed the more representative the measure was for the more ex-

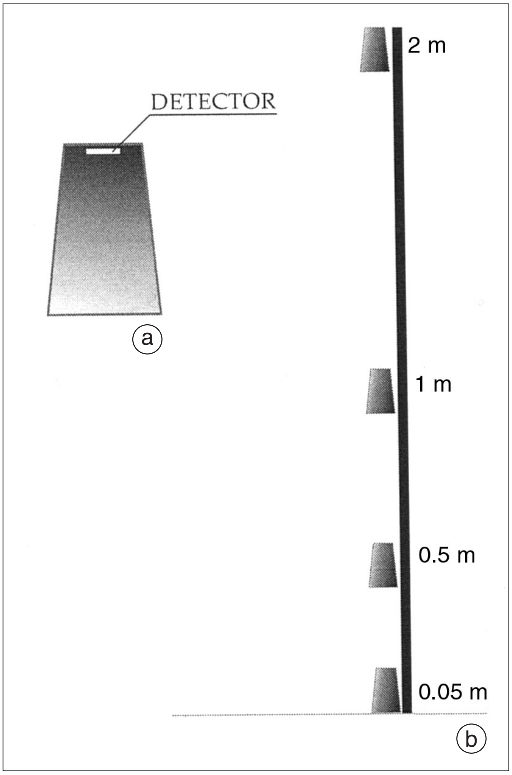

Fig. 2a,b. The measurements of radon activity in the outdoor air, a) a plastic cup with detector; b) diagram presenting the construction of the monitoring point.

tensive area. Furthermore, detectors situated 0.05m above the ground registered thoron Rn^{220} beside the radon Rn^{222}. The time of exposure was 6 months, twice a year: October-March (autumn-winter period) and April-September (spring-summer period). A long measurement period enables us to obtain the average values for the time of exposure and reduces the effect of diurnal fluctuations of pressure and temperature. The two sets of measurements were repeated the following year and the values obtained were similar to these which had been taken the year before.

The radon activity in the outdoor air was calculated dividing the number of traces on 1 cm^2 of the detector surface by the number of days of exposure and the result obtained multiplied by the calibration coefficient (in case of the atmospheric air it is 13.8), this model was elaborated by Srivastawa *et al.* (1995).

The LR-115 detector was also used during the measurement of the radon emanation from different types of rocks from the investigated area. To measure *the radon emanation* a modified method of the «can technique» was applied. The «can technique» was developed by Alter and Prize (1974 fide Azam *et al.*, 1995) and then applied with some modification, among others, by Karamadoust (1988), Azam *et al.* (1995) and Solecki (1999).

The basis of this method is that the total amount of radon in the investigated material consists of two parts, one – the atoms of radon stuck in the mineral crystals, the other – the radon atoms free to migrate in the interstitial spaces. The second part of radon atoms is the part which can get out of the rock material, enter the air above and could be registered by the detector. The ratio of these parts is defined by the emanation coefficient. The emanation coefficient was calculated introducing the total radium activity (content) $C_{t\mathrm{Ra}}$ and effective radium activity $C_{e\mathrm{Ra}}$

$$k_e = C_{e\mathrm{Ra}} / C_{t\mathrm{Ra}}.$$

The total radium activity $C_{t\mathrm{Ra}}$ is the total amount of radon in the rock material calculated on the basis of gamma-spectrometric analysis of radium activity. In this case the analysis was made by Radiometric Laboratory of GIG in Katowice, using semi-conductor detector HPGe, according to the Polish norm PN-89/Z-70073. The effective radium content $C_{e\mathrm{Ra}}$ is the fraction of radium which corresponds to the part of radon which has emanated from the sample. Effective radium activity is expressed in Bq/kg unit and can be calculated from the equation

$$C_{e\mathrm{Ra}} = \frac{\rho V}{K M T_e}$$

where ρ is the track density on cm^2 of the detector surface, V the volume of free space in the can, K a calibration constant (0.0245 track on cm^2 in one day), M the mass of sample in kg and T_e the time of exposure in days (Azam *et al.*, 1995).

Crushed rock material (grain size of 3.5-1 cm) was closed in a hermetic can of the known volume. On the inner surface of the cap, there was fixed a detector to register the alpha particles of radon in the air above the sample of rock (fig. 3). The important element is to start exposure after stabilization of the secular equilibrium between Ra^{226} and Rn^{222}. The equilibrium (of 98%) between them stabilizes after 3 weeks in the closed space. Because of this period of stabilization, 3 weeks after closing the can, the detector was covered by metal plate, which was held by a magnet from outside of the can. After this period the metal plate was removed by taking away the magnet and the detector starts to be exposed. This is a modification of the Azam method introduced by Solecki (1999). The detector mainly registers the alpha particles which come from the decay of Rn^{222} and not from Rn^{220}. Taking into account diffusion coefficients of radon in the air and in the crushed rock material, the half-life time of the thoron Rn^{220} and the distance between the detector and the surface of the sample, it was estimated that alpha particles of thoron could be registered in 10%. The emanation coefficients of 14 different types of rocks were measured and estimated. Moreover the emanation coefficient was measured for a grain size smaller than 1 cm and for a moisture content of 13% and 24% wt for each type of rock. These variations of conditions changed the k_e values but the relative differences of k_e between the different types of rocks remained the same. In this paper k_e values are presented which were obtained for dry rock material of grain size of 3.5-1 cm.

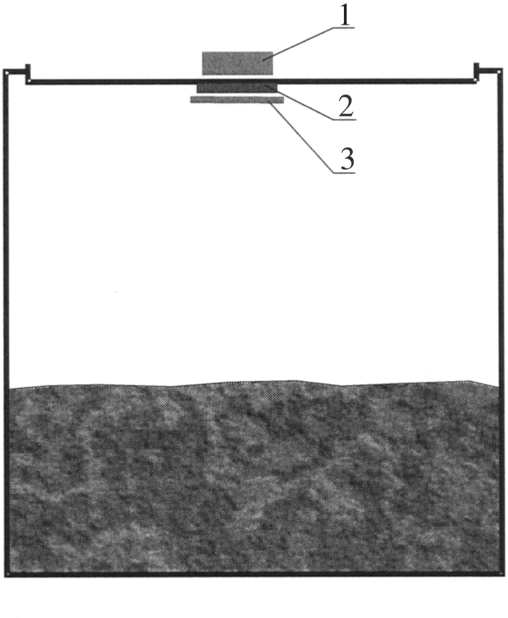

Fig. 3. The instrument for measuring radon emanation from the rock material by «can technique» method; 1 – magnet, 2 – detector, 3 – metal plate.

The field measurements of the *uranium and thorium content in rocks and soil* were performed using gamma-spectrometer GR-320. The gamma-spectrometer GR-320 is equipped with a detector, with a source Cs of 0.5 mCi (18.5 kBq). The gamma-spectrometer GR-320 measures contents of Bi^{214} and Tl^{208} and on this basis, it estimates the contents of U^{238} and Th^{232}. Therefore the results are presented as an equivalent content of uranium (eU) and thorium (eTh). A few zero values of measurements obtained on the outcrops of gabbro and serpentinites prove that the cosmic radiation does not influence the results of measurements.

The 29 sets of measurements (in each place – 30 single measurements, each one lasting 300s) were collected above the outcrops of different type of rocks and above the soils which cover them.

4. Results

4.1. *The eU and eTh content in soil and underlying rocks*

The mean eU contents in the rocks of investigated area vary from values close to zero (gabbro, serpentinite) up to 12 ppm, in the quartz-graphite schist in the south part of Niemcza Zone (fig. 4). The quartz-graphite schist which crops out in the north part of metamorphic of Niemcza-Kamieniec Ząbkowicki, contains 7 ppm eU. The mean values for granitoides are 3-6 ppm eU, in mica schist of metamorphic of Niemcza-Kamieniec Ząbkowicki and schist of Wzgórza Lipowe the measured contents are 2-4 ppm eU. A relatively high value was obtained for basalt in the north-east part of the metamorphic of Niem-

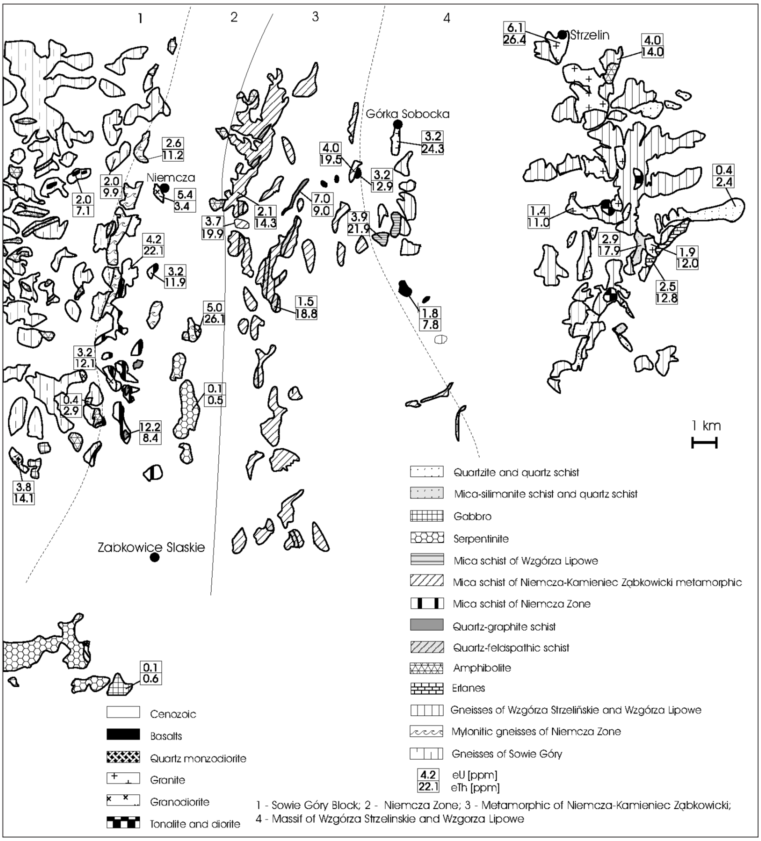

Fig. 4. The mean eU and eTh contents in the rocks of Wzgórza Niemczańsko-Strzelińskie.

cza-Kamieniec Ząbkowicki – 3.2 ppm. The lowest values of uranium content were measured on amphibolites, quartzite, quartz-feldspathic schist and some basalts.

The thorium content has a decisive impact the thoron activity in the environment. The relatively high eTh contents (> 20 ppm) were measured on the outcrops of some granite and schist of Wzgórza Lipowe (fig. 4). The highest mean value – 32 ppm, was found on granodiorite near Niemcza. The slightly lower thorium contents (20-15 ppm) were recorded in quartz-feldspathic schist of metamorphic of Niemcza-Kamieniec Ząbkowicki, mica schist of Wzgorza Lipowe and mica-sillimanite schist in south part of Wzgorza Strzelinskie. The high value eTh of the basalt in the north-east part of the metamorphic of Niemcza-Kamieniec Ząbkowicki – 12.9 ppm is noteworthy.

The uranium and thorium content in soil usually corresponds to the contents in the underlying rocks. In most places the eU and eTh contents in soil (mean values: 2 ppm eU and 8 ppm eTh) are slightly reduced in relation to the contents in underlying rocks (mean: 3 ppm eU and 12 ppm eTh), but not in every place. On the areas composed of amphibolite, serpentinite, gabbro and quartzite the values measured on soil are higher than the values measured on the underlying rocks. It could be explained by the presence of loess material in the soil, which contains, after Solecki (2000), 2-3 ppm eU and about 10 ppm eTh.

4.2. *The emanation coefficient of the samples of selected rocks*

Emanation coefficient (k_e) is interpreted as a ratio of the amount of radon that emanated to the air above the sample to the whole radon which was generated in the sample. Emanation coefficient is a feature of the material and quantifies the ability of this material to emanate radon-gas.

The emanation coefficient of samples of 14 selected rocks vary in the range 0.003-0.13 (table I). According to UNSCEAR, the emanation coefficient varies in range of 0.01 – 0.8 (1988 fide Robé and Labed, 1995; Markkanen and Arvela, 1992). The emanation coefficients of investigated rocks fit in the lower range of the values published by UNSCEAR. The lower emanation coefficients were measured for basalts and the highest values of emanation coefficient were obtained for quartz-feldspathic schist, quartz-graphite schist and granite (Górka Sobocka). Solecki (1999) measured for the metamorphic schist an emanation coefficient of 0.29. The higher emanation coefficient of metamorphic schist than that of the magmatic rocks could be explained by the textures of metamorphic schist, which were formed as a result of numerous transformations and deformations. Therefore it could be presumed that the migration inside these rocks will be faster than in the solid magmatic rock.

Table I. Emanation coefficient (k_e) and effective Ra^{226} activity of selected rocks from investigated area.

Type of rock	k_e	$C_e\mathrm{Ra}^{226}$ [Bq/kg]
Gabbro	0.01	0.03
Serpentine	0.03	0.13
Quartzite (Wzgórza Strzelińskie)	0.07	0.45
amphibolite (NE part of Sowie Góry)	0.06	0.80
Basalt (E periphery of Góry Sowie)	0.007	0.26
Basalt (metamorphic of Niemcza-Kamieniec Ząbkowicki)	0.003	0.17
Basalt (S part of Wzgórza Lipowe)	0.003	0.08
Quartz-feldspathic schist (metamorphic of Niemcza-Kamieniec Ząbkowicki)	0.13	3.39
Quartz-graphite schist (Niemcza Zone)	0.11	6.80
Gneisses (Wzgórza Strzelińskie)	0.01	0.55
Quartz monzodiorite (NE part of Sowie Gory)	0.01	0.51
Granodiorite (Niemcza Zone)	0.06	4.43
Granite (Górka Sobocka)	0.09	5.25
Granite (Strzelin)	0.03	1.90

Effective radium activity (content) ($C_e\mathrm{Ra}$) quantifies the radon that come out to the atmosphere from the definite quantity of crushed rock. Effective radium activity is the result of the total activity of Ra^{228} and Ra^{226} in rock and the emanation coefficient of this rock. The measurements inside the closed can partially include the thoron emanation, however in the atmosphere the presence of thoron is limited up to the height of 30-40 cm. Therefore in the case of atmospheric radon the effective Ra^{226} activity should by taken in consideration. The effective Ra^{226} activity for the investigated samples is shown in the last column of table I.

4.3. *Atmospheric radon activity in the air 2 m above the ground surface*

The radon activity distribution above the area of Wzgórza Niemczańsko-Strzelińskie is illustrated in fig. 5, the values are year-average. The highest radon activity, above 20 $\mathrm{Bq\ m}^3$, is observed above the area of the outcrops of mylonite of Niemcza Zone, where the maximum values were measured above the outcrops of mylonitic gneisses, granodiorite and quartz-graphite schist. Another area of high values is the region of Górka Sobocka, Wzgórza Lipowe and North-West part of Wzgórza Strzelińskie, with the maximum values above the outcrops of granites near Górka Sobocka and Strzelin. These two geological situations of the higher radon activity in the atmosphere demonstrate the two general factors

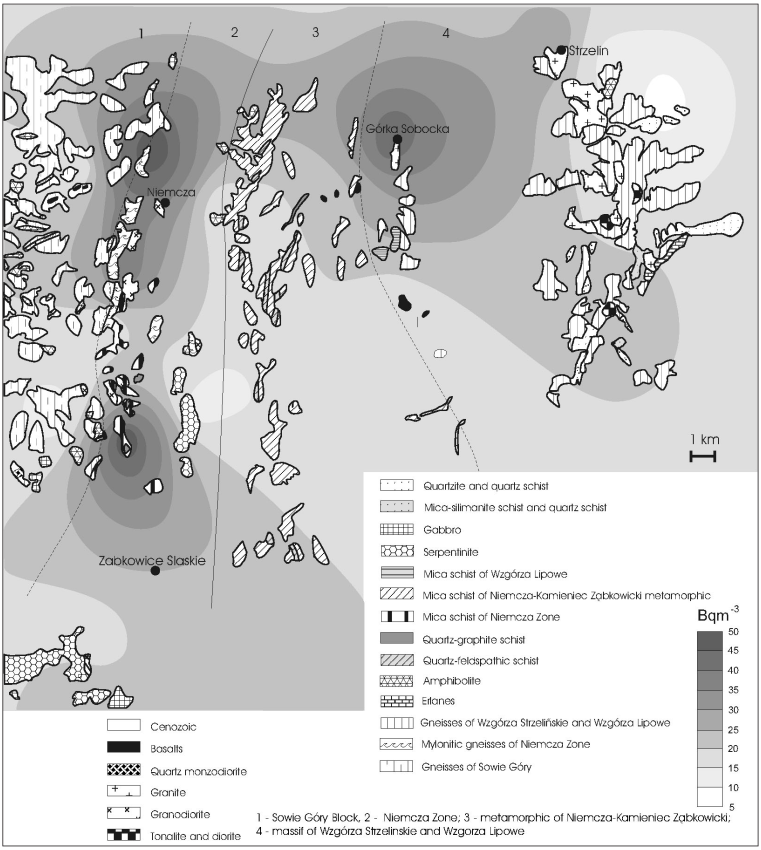

Fig. 5. Atmospheric radon activity distribution above the area of Wzgórza Niemczańsko-Strzelińskie.

that influence the radon exhalation: uranium/radium content in the rocks and their emanation coefficient. The influence of the high contents of uranium in rocks on the radon activity in the atmosphere is obvious. Much more interesting seems to be the influence of the mylonitisation, fracturating and weathering of rocks in the tectonic zone, which can be observed above the Niemcza Zone. The increased migration of radon from fractured and weathered rocks was described among others by Gates *et al.* (1990), Ball *et al.* (1991), Gundersen (1991), Dubois *et al.* (1995) and Ciężkowski and Przylibski (1997). Faults and cracks increase the surface of contact between rock and ground water, what promotes the possibility of radon migration too (Ball *et al.*, 1991; Strzelecki and Wolkowicz, 1993).

The mean value of atmospheric radon activity, measured 2 m above the ground, on the area of Wzgórza Niemczańsko-Strzelińskie was 21 Bqm^{-3}. This value corresponds to the radon activity measured above the central mountainous areas of the USA. The mean radon activity in the atmosphere on the area of USA varies in the range of 4-15 Bqm^{-3}, on the Colorado Plateau these values reach 18.5-27.8 Bqm^{-3} (Gesell, 1983). In Canada outdoor radon activity measured 3 m above the ground surface reached the mean values for the provinces of Manitoba 59 Bqm^{-3} and Saskatchewan 61 Bqm^{-3} (Grasty, 1991).

4.4. *Radon activity in the air close to ground, 0.05 m above the ground surface*

The year-average values of radon activity close to the ground are shown in table II. The values are not interpolated over the whole area because this parameter varies strongly, depending on the exhalation from the limited local ground. Moreover, because of the way the cups with detectors are positioned (they were placed directly on the ground), the measurements were not influenced by dispersion in the atmosphere. The measurements carried out close to the ground include Rn^{222} and Rn^{220}, so the radon activity values are influenced by both uranium and thorium content in soil and underlying rocks.

The highest values of radon activity (>200 Bqm^{-3}) in air close to the ground were measured on granodiorite and granite, a little lower values (100-200 Bqm^{-3}) were measured on mylonitic gneisses of Niemcza Zone, on mica schist and quartz-feldspathic schist of metamorphic of Niemcza-Kamieniec Ząbkowicki and on some basalts. The high values of radon activity are caused by the relatively high uranium and thorium content in soil (2-3 ppm eU and 7-18 ppm eTh) and underlying rocks (3-6 ppm eU and 11-26 ppm eTh) in these places. The lowest activities (25-45 Bqm^{-3}) were measured on serpentinite and gneisses of Góry Sowie, where the uranium and thorium contents are: close to zero for eU and 2-3 ppm eTh. The relatively low radon activity of 52 Bqm^{-3} on quartz-graphite schist, where the contents of eU and eTh in rock are 13 ppm and 6.6 ppm respectively, results from the unavoidable location of the detector outside of the old quarry, near the gneiss outcrops. The high values of radon activity in the air in relation to the low uranium and thorium contents in underlying rocks, were measured on amphibolites, gabbros and quartz. In these places the soil contains more radioactive elements (1.5-2 ppm eU and 6-9 ppm eTh) than the rocks (0.1-0.5 ppm eU and 0.7-3.7 ppm Th), which could result from the uranium and thorium concentration during the soil genesis process and from addition of loess material.

Table II. Radon activity in the air close to the ground above the outcrops of different rocks.

Number of monitoring point (fig. 1)	Type of underlying rock	Rn activity 0.05 m above the ground [Bqm^{-3}]
1	Mylonitic gneisses of Niemcza Zone	193
2	Basalt	171
3	Granodiorite	303
4	Mica schist of Niemcza Zone	90
5	Amphibolite	118
6	Quartz-graphite schist	52
7	Gneisses of Sowie Góry	25
8	Quartz monzodiorite	66
9	Serpentinite	45
10	Granodiorite	212
11	Serpentinite	37
12	Gabbro	85
13	Amphibolite	165
14	Mica schist	67
15	Quartz-feldspathic schist	131
16	Mica schist	137
17	Mica schist	180
18	Mica schist	155
19	Basalt	87
20	Basalt	101
21	Granite	240
22	Granite	200
23	Gneisses of Wzgórza Strzelińskie	48
24	Gneisses of Wzgórza Strzelińskie	47
25	Quartzite and quartz schist	40
26	Granite	145
27	Erlanes	75

4.5. *Seasonal variation of radon activity in the air*

It is possible to observe slight seasonal variations of radon activity in the air at a height of 2 m (fig. 6). In spite of the fact that the modal value is located in the same range of activity val-

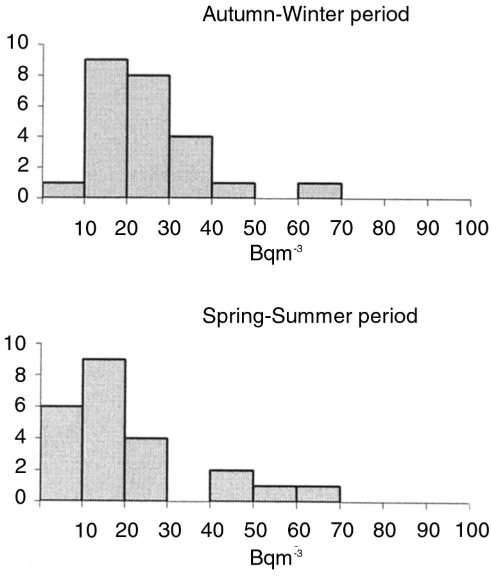

Fig. 6. Distribution of radon activity values, measured in the air 2 m above the ground surface in the two periods of the year, which demonstrates slight seasonal variation.

ues for the autumn-winter and spring-summer period (10-20 Bqm^{-3}), in the autumn-winter period there are more measurements in the range of 20-30 and 30-40 Bqm^{-3} than in the range of 0-10 Bqm^{-3}, the opposite situation is observed on the histogram for the spring-summer period.

The slightly higher values in the autumn-winter period could be explained by increased exhalation in the conditions of temperature difference between the air in the ground and air above the ground surface. In the autumn-winter period the air temperature above the ground usually is lower than that in the ground, which causes convection of the air from ground to the atmosphere. These phenomena were described by Wilkening (1990), Hakl *et al.* (1995) and Robé and Labed (1995). The snow cover, described as a factor which decreases the radon activity in the atmosphere (Juzdan *et al.*, 1985; Somogyj *et al.*, 1986; Feichter and Crutzen, 1989; Dörr and Münnich, 1990; Jacob and Prather, 1990; Ennemoser *et al.*, 1995), is not the dominant factor because on this area the period of snow cover is very short.

5. Conclusions

The mean value of radon activity in the air 2 m above ground surface was 21 Bqm^{-3}. The highest values were measured in the area of granite and quartz-graphite schist outcrops (rocks of the high eU content) and in the area of mylonitic rocks of the Niemcza Zone. These observations confirm that the radon activity in the atmosphere depends on uranium/radium content in the rocks and their emanation coefficient and on the mylonitisation and fracturating grade of rocks in the tectonic zone. The slight seasonal variation of radon activity in the air is the result of weather conditions which control radon migration from soil-gas to atmosphere.

Radon activity close to the ground surface varies from 25 to 300 Bqm^{-3} and accurately reflects the uranium and thorium content in the indirect ground basement (soil and weathered rocks).

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