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NATURAL RADIOACTIVITY OF ROCKS OCCURRING IN THE CONTACT ZONE OF THE KARKONOSZE MASSIF WITH THE SZKLARSKA PORĘBA SCHIST BELT

Aleksandra BIEDA * and Grzegorz LIZUREK

University of Silesia, Faculty of the Earth Sciences, Będzińska 60, 41-200 Sosnowiec, Poland,
Phone: (032) 3689511
**Corresponding author's e-mail:* abieda@wnoz.us.edu.pl

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ABSTRACT

The aim of the paper is to present the results of measurements of ^{40}K, ^{208}Tl, ^{212}Pb, ^{214}Pb, ^{214}Bi and ^{228}Ac activity concentrations of selected rocks collected from the contact zone of the Karkonosze massif with the Szklarska Poręba schist-belt. Activity concentrations were obtained using a gamma-ray spectrometry system. Determined activity concentrations of ^{40}K and nuclides from ^{238}U and ^{232}Th series in measured samples were compared with average activity concentrations of these radionuclides in the continental crust and in the soil and with data concerning investigated area available in the selected literature. In all rock samples very high activity concentrations associated with decay of ^{232}Th and ^{238}U series were observed. These values twice exceed the average activity concentrations refer to continental crust and are distinctly higher than activity concentrations measured in typical rocks of the Izera block.

KEYWORDS: radium, actinium, natural radioactivity, Karkonosze-Izera block, hornfels, granite, contact metamorphism

GEOLOGICAL SETTING

The study area is located in southwestern Poland, in the Izera Mountains (Western Sudetes) and covers the contact zone of the Karkonosze massif with the Szklarska Poręba schist-belt. Investigated area belongs to Karkonosze-Izera block (first-order geological unit in the Sudetes) which lies in the northern part of the Bohemian Massif. Szklarska Poręba schist-belt consists of cordierite-andalusite hornfelses and andalusite-cordierite hornfelses intercalated with amphibolites and laminated fine-augen gneisses (Żaba, 1979), (Cieśliński and Żaba, 1990). Hornfelses were formed due to contact metamorphism of the Proterozoic schists at the contact with Variscan granitoids intrusion (the Asturian phase). The contact metamorphism is both thermal and metasomatic and is found over a vast lateral range (Malczewski and Żaba, 2007).

Rock samples were collected from two places: closed pyrite mine located at the contact with the Karkonosze granite at Szklarska Poręba Średnia and the Zbójeckie Skały at Szklarska Poręba Dolna. In an open part of the pyrite mine cordierite-andalusite hornfelses were collected. From the second location 3 kinds of rock samples were taken: granite, hornfels, and the sample from the "contact" between the granite and the hornfels. The boundary between Karkonosze granite and the Izera block can be seen in this outcrop (granites underlie the hornfelses).

MATERIALS AND METHODS

All measurements were carried out in the Laboratory of Natural Radioactivity of Faculty of the Earth Sciences in Sosnowiec. The natural radioactivity of ^{40}K, ^{208}Tl, ^{212}Pb, ^{214}Pb, ^{214}Bi, and ^{228}Ac in collected rock samples were measured using the Canberra XtRa (Extended Range) HPGe detector model GX3020 (32% relative efficiency, crystal length: 58 mm, diameter: 50.5 mm). The energy resolutions of the detector equal 0.86 keV at 122 keV and 1.76 keV at 1.33 MeV.

This model of the detector was designed for *in situ* measurements but can be also used in laboratory conditions shielded with lead bricks to limit surroundings influence.

Each rock sample was crushed for homogenisation previously. Measurements were carried out in Marinelli 450 type geometry. Counting time was set at 48 h for each sample. The obtained spectra were analysed with the use of Canberra Genie 2000 software version 3.0. The determination of the presence of radionuclides and calculation of their activities were based on the following gamma-ray transitions (in keV): ^{40}K (1460.8), ^{208}Tl (583.1, 860.5, 2614,5), ^{212}Pb (238.6, 300.1), ^{214}Pb (241.9, 295.2, 351.9), ^{214}Bi (609.3, 1120.3, 1238.1, 1764.5), ^{228}Ac (338.3, 911.1, 968.9)

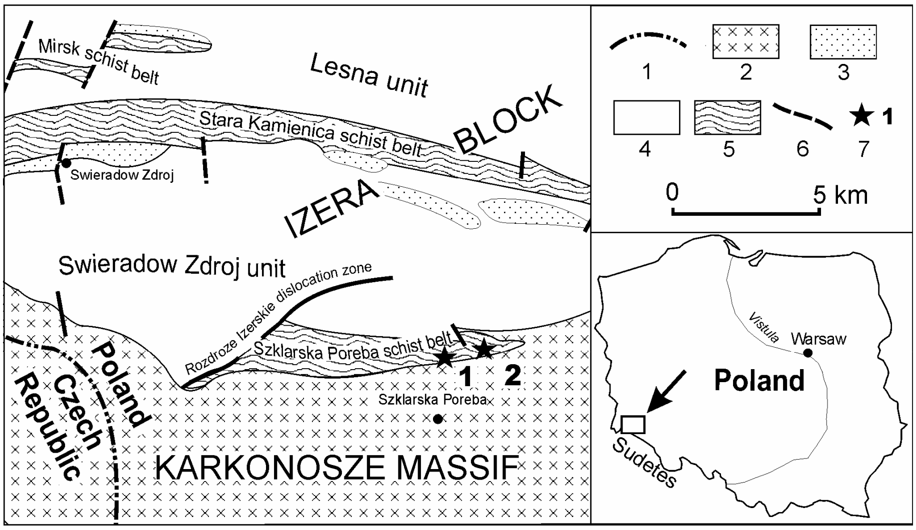

Fig. 1 Location of sampled places on the background of the Karkonosze-Izera block. 1-state border, 2-Karkonosze granite, 3-leucogranites, 4-Izera gneisses, granite-gneisses and granites, 5-mica-schists, 6-main faults, 7-sampled places (1-pyrite mine, 2-Zbojeckie Skaly).

Table 1 Measured activity concentrations of ^{40}K, ^{208}Tl, ^{212}Pb, ^{228}Ac, ^{214}Pb, ^{214}Bi with calculated ^{226}Ra activity concentrations and concentrations of K (%), ^{232}Th and ^{238}U (ppm) in investigated rock samples. Uncertainties are given within one standard deviation.

Nuclide	Activity concentration (Bq·kg^{-1})			
	Zbójeckie Skały			piryte mine
	granite	"contact"	hornfels	hornfels
K-40	1075 ± 23	1122 ± 24	1199 ± 26	1385 ± 30
Tl-208	34.8 ± 1.2	37.7 ± 1.3	36.4 ± 1.3	29.1 ± 1.0
Pb-212	102.7 ± 5.6	111.0 ± 6.0	109.2 ± 5.9	85.2 ± 4.6
Ac-228	107.3 ± 3.7	111.9 ± 3.8	108.3 ± 3.7	86.0 ± 2.9
Pb-214	72.1 ± 3.8	87.1 ± 4.6	79.5 ± 4.2	50.5 ± 2.7
Bi-214	56.3 ± 2.0	69.6 ± 2.4	63.0 ± 2.2	38.3 ± 1.6
Ra-226	64.2 ± 2.9	78.3 ± 3.5	71.2 ± 3.2	44.4 ± 2.1
	Concentration			
K (%)	3.5 ± 0.1	3.7 ± 0.1	4.0 ± 0.1	4.6 ± 0.1
Th-232 (ppm)	26.1 ± 0.9	27.2 ± 0.9	26.3 ± 0.9	20.9 ± 0.7
U-238 (ppm)	5.2 ± 0.2	6.3 ± 0.3	5.7 ± 0.3	3.6 ± 0.2
Th/U	5.1	4.3	4.6	5.9

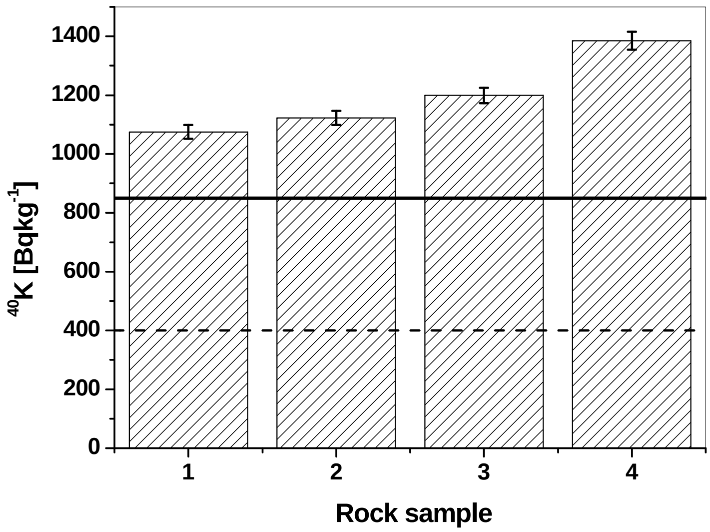

Fig. 2 Comparison of ^{40}K activity concentrations in measured rock samples with average activity concentrations of this radionuclide reported for the continental crust (solid line) and for the soil (dashed line). Samples 1, 2 and 3 - granite, sample from the contact between granite and hornfels and hornfels, respectively (Zbójeckie Skały), sample 4 – hornfels from closed pyrite mine.

RESULTS AND DISCUSSION

In the upper part of Table 1 measured activity concentrations of ^{40}K, nuclides from ^{232}Th series (^{208}Tl, ^{212}Pb, ^{228}Ac) and ^{238}U series (^{214}Pb, ^{214}Bi with ^{226}Ra) in investigated rock samples are presented. ^{226}Ra activity concentrations were calculated as the arithmetic means of activities of ^{214}Pb and ^{214}Bi isotopes. In the lower part of the table concentrations of K (%), ^{232}Th and ^{238}U (ppm) in measured samples calculated using conversion factors given by Polish Central Laboratory for Radiological Protection are showed. The ^{232}Th and ^{238}U concentrations are based on the ^{228}Ac and ^{226}Ra activity concentrations respectively. For all rocks Th/U ratio was calculated.

^{40}K

As shown in Table 1 ^{40}K activity concentrations for all rock samples are greater than 1000 Bq·kg^{-1}. In samples from the Zbójeckie Skały activity concentrations of this radionuclide don't differ much, but the lowest value was noted in granite (1075 ± 23 Bq·kg^{-1}) whereas the medium and the highest value refers to the sample collected from the contact between granite and hornfels (1122 ± 24 Bq·kg^{-1}) and to hornfels (1199 ± 26 Bq·kg^{-1}) respectively. In the sample taken from the pyrite mine activity concentration of ^{40}K is higher (1385 ± 30 Bq·kg^{-1}). As measurements show there can be a relation between content of ^{40}K in rocks and the distance of sampled places from granitoids intrusion (the farther from the intrusion the highest activity of potassium in rocks), but it's necessary to check concentration of this isotope in more locations. Concentration of potassium in investigated rocks varies in the range from 3.5 % (granite) to 4.6 % (hornfels from pyrite mine).

Obtained results were compared with average activity concentrations of ^{40}K reported for the continental crust i.e. 850 Bq·kg^{-1} and for the soil i.e. 400 Bq·kg^{-1} (Eisenbud and Gesell, 1997) and presented in Figure 2. As can be seen measured activity concentrations associated with ^{40}K decay are distinctly higher than the average activity concentration of this isotope in the continental crust for all samples.

^{232}Th

Data presented in Table 1 show that radioactive equilibrium between progenies in ^{232}Th series for all rock samples can be assumed. Measured rocks are characterized by very high values of activity concentrations of ^{228}Ac (^{232}Th). In samples collected from the Zbójeckie Skały those values are at the same level (about 109 Bq·kg^{-1}) and the higher value refers to the sample from the contact between granite and hornfels. In hornfels taken from the pyrite mine activity concentration of ^{228}Ac (^{232}Th) is distinctly lower (86.0 ± 2.9 Bq·kg^{-1}). Concentration of ^{232}Th in investigated rocks varies in the range from 20.9 ppm (hornfels from pyrite mine) to 27.2 ppm (sample from the "contact").

Noted activity concentrations are compared with average activity concentrations of ^{232}Th in the

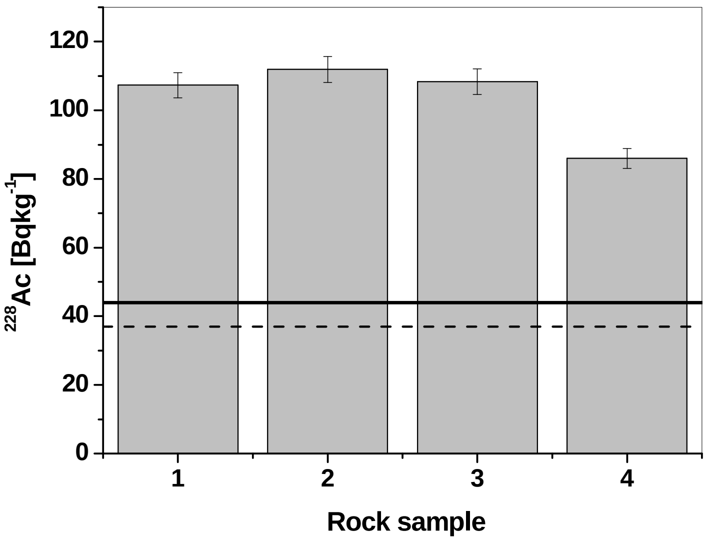

Fig. 3 Comparison of ^{228}Ac (^{232}Th) activity concentrations in measured rock samples with average activity concentrations of ^{232}Th reported for the continental crust (solid line) and for the soil (dashed line). Samples 1, 2 and 3 - granite, sample from the contact between granite and hornfels and hornfels, respectively (Zbójeckie Skały), sample 4 – hornfels from closed pyrite mine.

continental crust i.e. 44 Bq·kg^{-1} and in the soil i.e. 37 Bq·kg^{-1} (Eisenbud and Gesell, 1997) in Figure 3. Even the lower measured value of ^{228}Ac (^{232}Th) activity concentration (hornfels from pyrite mine) exceeds the average activity concentration refers to the continental crust almost twice.

^{38}U

Measured rocks are characterized by high radioactivity of ^{226}Ra (^{238}U). The highest values refer to the sample collected from the contact between granite and hornfels, (78.3 ± 3.5 Bq·kg^{-1}), lower values were obtained for hornfels (71.2 ± 3.2 Bq·kg^{-1}) and granite (64.2 ± 2.9 Bq·kg^{-1}). Distinctly lower value was noted in hornfels from the pyrite mine (44.4 ± 2.1 Bq·kg^{-1}). For comparison arithmetic mean of activity concentrations of ^{226}Ra in hornfelses from Lower Silesia equals 35.7 Bq·kg^{-1}. ^{226}Ra activity concentrations in granites from Szklarska Poręba (Karkonosze granite) vary in the wide range from 15 to 119 Bq·kg^{-1} whereas in aplite and in mica schist from Szklarska Poręba ^{226}Ra activity concentrations equal 66 ± 6 Bq·kg^{-1} and 54 ± 9 Bq·kg^{-1} respectively (Przylibski, 2004).

Concentration of ^{238}U in investigated rocks calculated with assumption of radioactivity equilibrium in uranium series varies in the range from 3.6 ppm (hornfels collected from the pyrite mine) to 6.3 ppm (sample taken from the “contact”).

Measured activity concentrations of ^{226}Ra (^{238}U) were compared with average activity concentrations of ^{238}U reported for the continental crust i.e. 36 Bq·kg^{-1} and for the soil i.e. 22 Bq·kg^{-1} (Eisenbud and Gesell, 1997) and they are presented in Figure 4. For each case the average activity concentrations of ^{238}U in the crust is exceeded (for the sample taken from the “contact” over twice).

Th/U

As it was mentioned above for all rock samples Th/U ratio was calculated. This ratio varies in the range from 4.3 to 5.9. The highest value refers to the hornfels from the pyrite mine, whereas the lowest value was noted in the sample collected from the contact between granite and hornfels. In granite this ratio equals 5.1 in hornfels 4.6 respectively.

Obtained Th/U concentration ratios are very high, strongly higher than data published in literature concerning rocks of Karkonosze-Izera block. For example, in hornfels from the Death Bend area Th/U equals 3 and Th/U concentration ratio in rocks in the environs of Świeradów Zdrój varies between 1.5 and 3.2 (Malczewski et al., 2004), Th/U in selected crystalline rocks of the Izera block varies between 0.8 and 2.4 (Malczewski et al., 2005). It must to be noted that mentioned above results from the literature were obtained at the basis of in situ measurements (using a portable gamma-ray spectrometry workstation EG&G ORTEC). In field conditions detector view for gamma emitters is about 10 m in radius to a depth of 15-30 cm. In this case influence of the environment is a fundamental factor causing that results of in situ

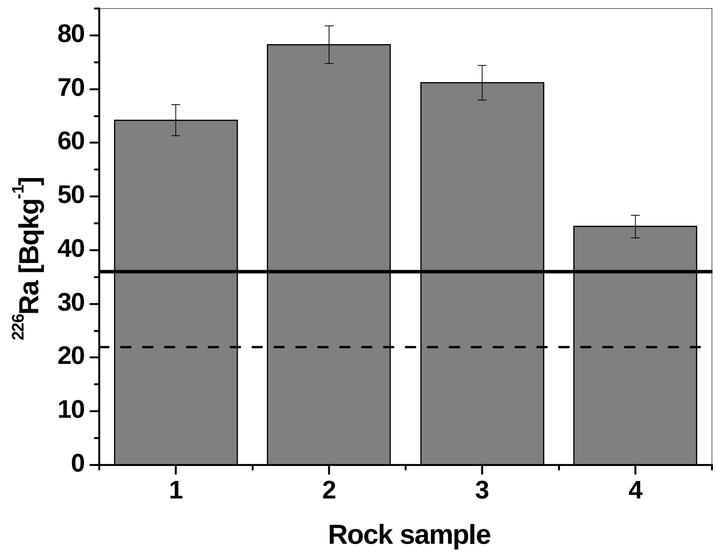

Fig. 4 Comparison of ^{226}Ra (^{238}U) activity concentrations in measured rock samples with average activity concentrations of ^{238}U reported for the continental crust (solid line) and for the soil (dashed line). Samples 1, 2 and 3 - granite, sample from the contact between granite and hornfels and hornfels, respectively (Zbójeckie Skały), sample 4 – hornfels from closed pyrite mine.

measurements may give lower values of radionuclides activity concentrations that laboratory measurements.

Th/U concentration ratio given by Eisenbud and Gesell (1997) for the continental crust equals 1.2 and for the granites is 1.8. Cited values are distinctly lower than Th/U concentration ratio obtained for investigated rock samples collected from the contact zone of the Karkonosze massif with the Szklarska Poręba schist-belt. Even value of Th/U ratio measured in granites from Szklarska Poręba that equals 3.2 (Plewa and Plewa, 1992) is lower.

Obtained results suggest that very high content of ^{228}Ac (^{232}Th) and high content of ^{226}Ra (^{238}U) in investigated rock samples are connected with specific collection place, where rocks formed due to thermal metamorphism and metasomatic processes occur. These processes seem to be responsible for so high level of radioactivity of rocks but it needs further geochemical research. The highest value of Th/U concentration ratio refers to the hornfels from the pyrite mine, which is the farthest location from the contact with the Karkonosze granite. The lowest values of ^{228}Ac (^{232}Th) and ^{226}Ra (^{238}U) activity concentration were measured in rock sample collected from this site. In this case the lowest difference between concentrations of ^{232}Th and ^{238}U among the other rock samples is observed.

COMPARISON OF OBTAINED RESULTS FOR THE ROCK SAMPLES WITH K (%), ^{232}Th AND ^{238}U (ppm) CONCENTRATIONS IN SOILS

Obtained concentrations of K (%), ^{232}Th and ^{238}U (ppm) in measured rock samples were compared with the newest results concerning natural radioactivity of the soils occurring in the investigated area (Malczewski and Żaba, 2007). Both rock and soil samples were collected from the same places and measured in the same laboratory using the same detector.

In Figure 5 comparison of K (%), ^{232}Th and ^{238}U (ppm) concentrations in rock and in soil samples collected from the Zbójeckie Skały is presented. Concentrations of these radionuclides in rock were calculated as the arithmetic mean of concentrations of K (%), ^{232}Th and ^{238}U (ppm) in three rock samples: granite, sample collected from the contact between granite and hornfels and hornfels.

Figure 6 shows comparison of K (%), ^{232}Th and ^{238}U (ppm) concentrations in hornfels and in soil developed on hornfels collected from closed pyrite mine.

As can be seen in the Figure 5 and Figure 6 calculated concentrations of radionuclides in investigated rocks are different from the concentrations of the same radionuclides in soil

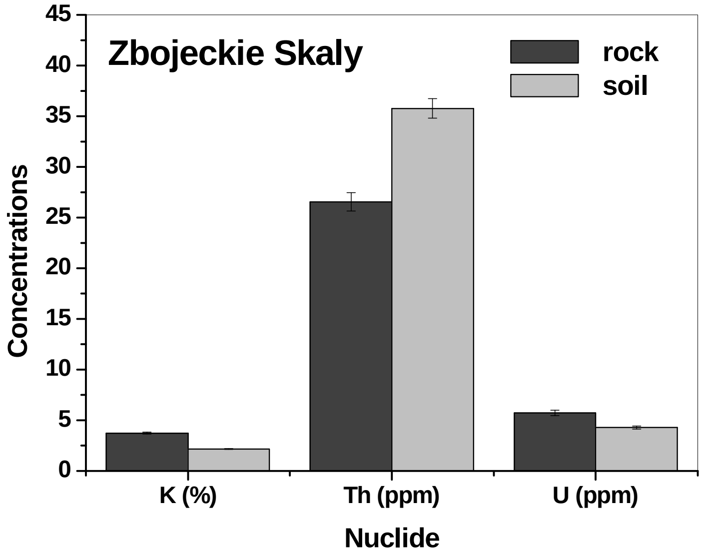

Fig. 5 Comparison of K (%), ^{232}Th and ^{238}U (ppm) concentrations in rock and in soil samples collected from the Zbójeckie Skały. Dark bars represent concentrations of K (%), ^{232}Th and ^{238}U (ppm) in measured rock samples and the light grey bars represent concentrations of these radionuclides in soil.

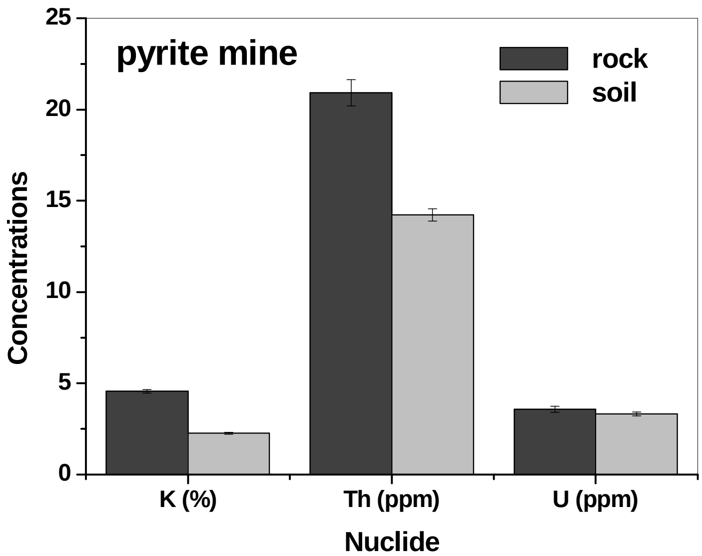

Fig. 6 Comparison of K (%), ^{232}Th and ^{238}U (ppm) concentrations in hornfels and in soil developed on hornfels collected from closed pyrite mine. Dark bars represent concentrations of K (%), ^{232}Th and ^{238}U (ppm) in measured rock sample and the light grey bars represent concentrations of these radionuclides in soil.

collected from the same location, but they both show the general information about the radioactivity of investigated material: samples are characterized by very high concentrations of thorium and uranium and high concentration of potassium.

Obtained values confirm cited results despite measured material are different (rock and soil).

CONCLUSIONS

The results of gamma-ray measurements presented in the paper give current information about natural radioactivity of rock samples collected from the contact zone of the Karkonosze massif with the Szklarska Poręba schist-belt.

As obtained results show Variscan granite intrusion could determined high radioactivity of rocks formed due to thermal metamorphism. There can be a relation in measured rock samples between content of ^{40}K in rocks (hornfels) and the distance of sampled places from granitoids intrusion (the farther from the intrusion the highest activity of potassium in rocks) but it needs further measurements (in other locations) and geochemical research. In all rock samples very high activity concentrations associated with decay of ^{232}Th and ^{238}U series were observed. These values twice exceed the average activity concentrations refer to continental crust and are distinctly higher than activity concentrations measured in typical rocks of the Karkonosze-Izera block. On the base of our measurements it could be noted that concentration of ^{232}Th and ^{238}U in measured rocks depends on the distance between sampled place and the contact between Karkonosze granite and the Izera block. Significantly higher concentration of ^{232}Th and ^{238}U occur in samples from the Zbójeckie Skały where contact between Karkonosze granite and the Izera block is located. The highest activities of ^{228}Ac (^{232}Th) and ^{226}Ra (^{238}U) in samples from Zbójeckie Skały were noted in the sample collected from the „contact" between granite and hornfels, so the majority of radionuclides genetically connected with granite intrusion are built into metamorphosed rocks in the closest vicinity of the intrusion but it should be checked with geochemical investigations.

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