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Analysis of influence of environmental conditions and natural gamma radiation for radon concentration measurements

The influence of environmental conditions and natural gamma radiation of rocks nearby measurement points has been introduced in this paper to radon concentration measurements in soil air. Our studies helped to assess anomalies caused by major tectonic zones that occurring in the Carpathians. These anomalies, can be used for interpretation and location of potential zones for oil and gas industry. Environmental tests had to be conducted because of the introduction of new measurement methodology within evaluation of statistical measurement errors and specific tests conditions. Stationary work was carried out in a limited area nearby Bagrowa st., Krakow, fieldwork was carried out at the Cergowa Mountain–Krosno profile. The conducted research did not show the influence of air humidity, precipitation, air temperature, air pressure and natural radioactive elements (K-40, U-238 and Th-232) on the measured radon anomalies. In this study RAD7 spectrometer with a soil probe was used.

Key words: radon, environmental conditions, gamma radiation, spectrometry.

Analiza wpływu warunków środowiskowych oraz naturalnej promieniotwórczości gamma na wyniki pomiarów koncentracji radonu

W pracy przedstawiono wpływ warunków środowiskowych i naturalnej promieniotwórczości skał występujących w otoczeniu punktów pomiarowych, na wyniki badań koncentracji radonu w powietrzu glebowym. Badania radonu pozwalają na identyfikację anomalii powodowanych występowaniem głównych stref uskokowych w Karpatach, co może pomóc wyznaczyć lokalizację występowania obszarów o potencjale dla poszukiwań przemysłu naftowego. Konieczność przeprowadzenia testów związana jest z opracowaniem nowej metodyki, która wymagała oceny błędów pomiarowych oraz warunków, w jakich należy prowadzić badania. Prace stacjonarne wykonane zostały w obrębie ograniczonej powierzchni w Krakowie przy ul. Bagrowej, prace terenowe przeprowadzono na profilu Góra Cergowa–Krosno. Badania nie ukazały wpływu wilgotności powietrza, opadów atmosferycznych, temperatury powietrza, ciśnienia atmosferycznego oraz naturalnych pierwiastków promieniotwórczych (K-40, U-238, Th-232) na mierzone anomalie radonowe. Do przeprowadzenia badań wykorzystano aparaturę badawczą w postaci spektrometru RAD7 wraz z sondą do pomiarów gazu w powietrzu glebowym.

Słowa kluczowe: radon, warunki środowiskowe, promieniowanie gamma, spektrometria.

Introduction

The studies on the radon concentration in the soil air are carried out by means of a proven method used in a broad range of geological and exploration work. Numerous papers on the surface measurements of anomalous amounts of radon and other gases in the soil air confirm the possibility of unambiguous assessment of the main tectonic zones direction [22, 29]. Anomalous properties occurring above the fault zones are easy to identify and frequently described in the literature, while their classification is much more difficult and requires additional studies in the area of precisely documented structures [25]. Publications from the field of Earth sciences describe the application of radon measurement results in: determination of deep structures (faults) [10, 18, 21], exploration of metal ores, geothermal sources, in the oil industry [8, 9], earthquake prediction and in the monitoring of volcanoes activity [2]. Measurements of radon in fault zones were carried out also in the area of Poland, for example nearby Krakow agglomeration [26] and in the Sudetes Mountains, where e.g. in Kowary the highest concentrations in the country were registered [6, 16]. However, the measurements of radioactive gas concentration are most often carried out in dwellings due to its impact on the human health [13, 17].

A small degree of Carpathian profiles exposure forces to apply various methods (apart from cartographic) allowing to explore and determine the hierarchy of importance as well as the direction of tectonic zones, in particular those of open (unsealed) nature, predisposed to be the main migration routes [12, 14]. Among many gases existing in the sub-base (e.g. ^{4}He, CO_2, CH_4, ^{222}Rn) the radioactive radon-222 is most frequently applied in the exploration work due to well developed measuring techniques [18]. The radon origin in soils is related primarily to a high natural radioactivity of rocks composing lower part of the Earth core. The gas migrates from deeper situated strata through fracture zones to the surface. Owing to this, it is possible to track large fault zones and to monitoring the radon concentration variability in the superficial layers [8].

Radon-222 is a radioactive noble gas with a half-life of 3.8 days, existing in the radioactive series of uranium-238, which is a common component of magma, volcanic, metamorphic, and sedimentary rocks [24]. Radon-222 originates from the decay of radium-226, emitting the α radiation. During the decay, a newly originated radon atom is recoiled at a distance of a few nanometres, which results in trapping a part of atoms in grains, and another part migrates to the water or gas phase existing in the pore space. The decay products move towards the Earth's surface through diffusion and advection; in contact with the atmospheric air they are immediately dispersed [31]. The amount of radioactive gas generated in the pore space depends on the state of medium and on the grain size [28]. Radon migrating to the pore space constitutes 10 to 50% of this element total amount, the remaining part is trapped in the rock matrix. The range and the number of transported particles depend on the medium, in which they are retarded [20].

The estimation of the measurement uncertainty results is the necessity to analyse all phenomena, that can affect the obtained results. According to the literature data the measurements of radon concentration in the soil air can be affected by the weather conditions (e.g. atmospheric pressure fluctuations, rainfalls) [5, 15, 19, 25, 27]. The natural radioactivity of rocks existing in the immediate vicinity of measurement points is the second important factor to be considered.

Methods and techniques

The active alpha particle detection method [1] was used for the radon studies due to the performance of a large number of field measurements at a limited time. The work performed for this project was carried out using the following measuring instruments: a Durridge RAD7 radon detector with a probe for the soil air measurements, an LB-575/THP thermo-hygrobarometer, a Garmin 64s GPS/GLONASS receiver, and a GT-40 spectrometer for the natural radiation measurements.

The Durridge RAD7 radon detector is an instrument, which can be the basic unit of expanded measuring systems. Appropriate adaptation of settings allows to carry out tests in many different modes [4]. The instrument is equipped with a solid silicon semiconductor detector, converting the emitted alpha radiation into an electric signal. It is situated in the centre of a hemispherical sampling chamber of 0.7 litre capacity, which is coated inside with an electrical conductor. The internal high voltage power circuit charges the conductor to the voltage of 2000÷2500 V against the detector, creating an electric field in the chamber volume, attracting positively charged particles to the detector. An advantage of semiconductor detectors is their durability, which is important at carrying out the fieldwork, as well as their high resolution.

Radioactive isotopes emit alpha particles of characteristic energies, which are distinguished due to the signal strength. To determine the radon and thoron concentration the utilised 'SNIFF' mode uses only the signal originating from polonium-218 and polonium-216. The other products of decay are not considered. Because of the applied method the instrument enables the registration of frequent radon concentration changes and fast recovery after reaching high concentrations. When the short-lived polonium-218 decays on the active part of the detector, the probability of exciting an electric signal by an alpha particle originating from this decay is 50%. The subsequent decays produce *inter alia* the beta radiation, which is not registered [4].

The analysed spectrum is presented in an energy scale from 0 to 10 MeV (Figure 1). It presents an example of summary spectrum comprising a series of measurements. The peak of 5.3 MeV energy results from the lead-210 absorption. It does not affect the measurements quality, because it is not taken into account in the spectral analysis.

The counts in window A provide the information about the new radon concentration. When peaks in windows A and C have the same height, radon is in equilibrium – such graph

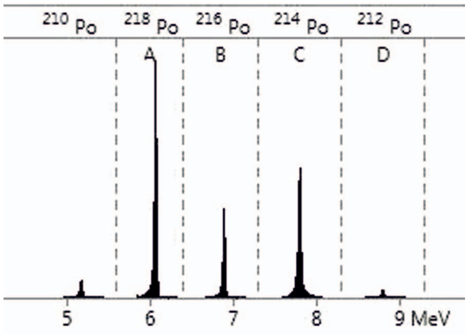

Fig. 1. Spectrum of registered counts for a series of measurements

type originates after a longer measurement. After purging the instrument's chamber (approximately about 10 minutes) the peak in window A disappears immediately, while the peak in window C is still visible. Windows B and D are responsible for measurements of thoron concentration, neglected in this paper [4].

A probe to carry out measurements in the soil profile is a separate element. Up to a depth of one metre the simplest probe is used, consisting of a sharpened tip with inlet openings, of a water-stopping valve and a vacuum gauge. The probe is connected with the RAD7 instrument by means of vinyl hoses. Appropriate filters and drying tubes are situated in the gas flow path. The paper made by Malczewski [16] presents changes of radon concentration in the soil air with the increasing depth of measurement. A significant increase in the concentrations, as against measurements at depths of 0, 10, and 40 cm, was shown for a depth of 80 cm. In the case of too shallow boreholes the soil air mixes with the atmospheric air, resulting in low concentrations. Hence studies at a depth of about 75 cm have been planned in this research [3].

The GT-40 spectrometer is an instrument for measurements of natural radioactive elements content (K-40, U-238, Th-232, and Cs-137). It can be used for environmental and rock exposure measurements, because it has built-in calibration used for studies of flat surfaces, minimum one metre in diameter. The spectrometer is built based on a two-inch BGO ($Bi_4Ge_3O_{12}$) detector, which allows to perform accurate measurements in a short time. The measurement time was set at 5 minutes per point.

A portable thermo-hygro-barometer made by the LAB-EL was used to monitor the weather conditions, providing the information about temperature, air humidity, and atmospheric pressure. It has an integrated memory, which allows to create the measurement history and to fulfil a recorder's function. The applied model calculates the current result of measurement based on the knowledge of non-linear characteristics of the temperature and humidity sensor. The range of measured temperatures comprises –40 to 85°C, the humidity range from 10 to 95% (for temperatures up to +60°C), the atmospheric pressure range is 500 to 1100 hPa [30].

Results and discussion

The studies aimed to determine the optimum methodology of measurements and to estimate the errors were carried out using the Durridge air probe at various arrangements and recording times (5 and 10 minutes, respectively). Tests were carried out in the area of INiG – PIB in Bagrowa 1, Krakow, within the area of approximate 2 metres square. The measuring mode was set to 'SNIFF' with an active thoron measurement option, in which the pump is turned on during the entire run. The results of studies are specified in Table 1.

For series 1 and 2, in which the measurements were carried out in one borehole without taking the probe out, an observation was made that the doubling of measurement time resulted in reducing the error three times, from 62.3 Bq/m^3 (45 minutes of measurement) to 21.5 Bq/m^3 (90 minutes of measurement). However, in the case of fieldwork it is not possible to perform that long analyses at one point. The measurement time was chosen so, that the measurement errors would not exceed the measured value.

In the next tests the measurements were performed in the area of approximate 2 m^2, after which the probe was taken out from the borehole and the measurements were repeated. Low errors (115.5 Bq/m^3) were obtained in series 3, in which the shortest purge and measurement time was set; it was affected by the averaged results of preceding measurements. A longer measurement time set in series 4 has substantially increased errors, to 281.8 Bq/m^3, which resulted from a too short period of chamber purifying. The elongation of the measurement time and the purge time to 10 minutes in series 5 allowed to reduce errors. In the case of series 6 the probe was additionally disconnected from the instrument, for a faster chamber recovery, which reduced errors to 146.1 Bq/m^3; unfortunately this result is still not satisfactory. The measurement time set to 10 minutes is a limit, in the case of making a few hundred measurements, is relatively long. More effective than measurement time extension would be extension of the chamber purging to 15 minutes, because this is approximate time between move to the next measurement point in the field.

In series 7 and 8 the measurement time was set to 10 minutes, and the purge time to 15 minutes. This substantially reduced the measurement errors to 66.1÷74.7 Bq/m^3. Results of those analyses were recorded during favourable weather conditions (7) and after the end of rainfalls (8). A significant difference in the

Table 1. Results of radon measurements (in Bq/m³ unit) for various arrangements and measurement times

1			2			3			4			5			6			7			8		
[Bq/m³]																							
Cycle time 5 min			Cycle time 10 min			Cycle time 5 min			Cycle time 10 min			Cycle time 10 min			Cycle time 10 min			Cycle time 10 min			Cycle time 10 min		
–			–			Purge 4 min			Purge 4 min			Purge 10 min			Purge 10 min			Purge 15 min			Purge 15 min		
505	±	369	687	±	251	1160	±	525	1860	±	411	1660	±	372	1330	±	335	220	±	189	2260	±	437
842	±	424	689	±	251	1020	±	468	842	±	316	387	±	199	1600	±	370	509	±	241	2340	±	444
977	±	442	637	±	243	1330	±	530	775	±	292	574	±	242	730	±	269	286	±	236	2110	±	433
640	±	390	536	±	226	1600	±	554	1630	±	398	1820	±	395	525	±	228	579	±	265	2050	±	425
1010	±	472	621	±	249	1310	±	509	1150	±	348	473	±	234	919	±	289	475	±	232	1920	±	430
842	±	43	621	±	249	745	±	400	3240	±	530	1760	±	390	559	±	237	203	±	196	–	–	–
741	±	430	655	±	254	885	±	441	2060	±	458	866	±	295	338	±	189	mean	**379**		mean	**2136**	
707	±	404	709	±	263	1150	±	496	2830	±	508	917	±	314	473	±	208	st. er.	**66.1**		st. er.	**74.7**	
1080	±	483	762	±	273	440	±	363	2120	±	442	1460	±	366	406	±	213						
mean	**816**		mean	**657**		mean	**1071**		mean	**1834**		mean	**1102**		mean	**764**							
st. er.	**62.3**		st. er.	**21.5**		st. er.	**115.5**		st. er.	**281.8**		st. er.	**192.3**		st. er.	**146.1**							

1, 2 – series of measurements in one borehole (continuous measurement).
3, 4, 5 – series of measurements in the area of approximate 2 m². Between measurements the probe was taken out from the borehole and the system was purged.
6, 7 – series of measurements in the area of approximate 2 m². Between measurements the probe was taken out from the borehole, disconnected from the system, and the chamber with the dryer was purged.
8 – series of measurements after a rainfall, in an arrangement similar to item 6 and 7.

registered values can be seen comparing both series, nevertheless the values overvalued in a similar way occurred also in series 4, which was carried out under conditions similar to series 7. For all measurements, in the cases when the probe was moved to a new borehole, the statistics was much poorer than in the case of performing continuous measurements at one point.

The waiting time for the detector purify does not have to be always the same. It depends on the previously recorded value. To estimate it, a test was carried out after a high radon value detection (Figure 2), which shows the reduction of activity in the chamber to below 1000 Bq/m³ after 24 minutes. Measurements after 2 and 4 minutes recorded higher values than the initial measurement (gas flow delay), because the gas analysis

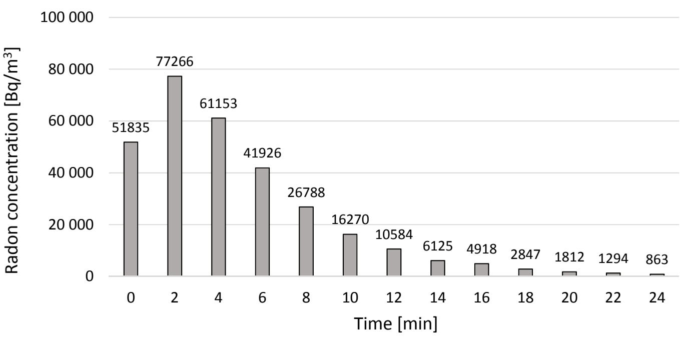

Fig. 2. Radon concentration graph, in function of time after the registration of a high concentration. Measurement was carried out during detection chamber purification

was made in an already filled chamber. The inspection of the measuring chamber cleaning level is very important and it should be monitored.

The air humidity, precipitation, air temperature, and the atmospheric pressure are the factors that can affect the results of radon measurements in the soil air. The precipitation seems to be the most important for the recorded parameters, because it directly affects the soil permeability. During the rainfall the ground surface is partly sealed with liquids, which slow down the gas permeation to the air. In such time particles of radioactive gas accumulate under the surface, which should increase the registered concentrations. The impact of precipitation on measurement results was recorded during stationary measurements.

The air temperature and the atmospheric pressure also control the movement of gases in the atmosphere and at its contact with the pedosphere. Depending on their values the gas faster or slower gets out from the soil layers. Moreno [19] registered seasonal changes of radon concentration and of air temperature during a year, showing a correlation of $r^2 = 0.66$. Those studies have shown much higher concentrations during the summer period. Also Tchorz-Trzciakiewicz [27] has shown the influence of seasonal conditions in the area of Poland, studying changes of radon concentration in a coal mine.

Measurements carried out for this paper were performed during the summer period at a similar air temperature. The air humidity featured the highest daily variations. The studies on

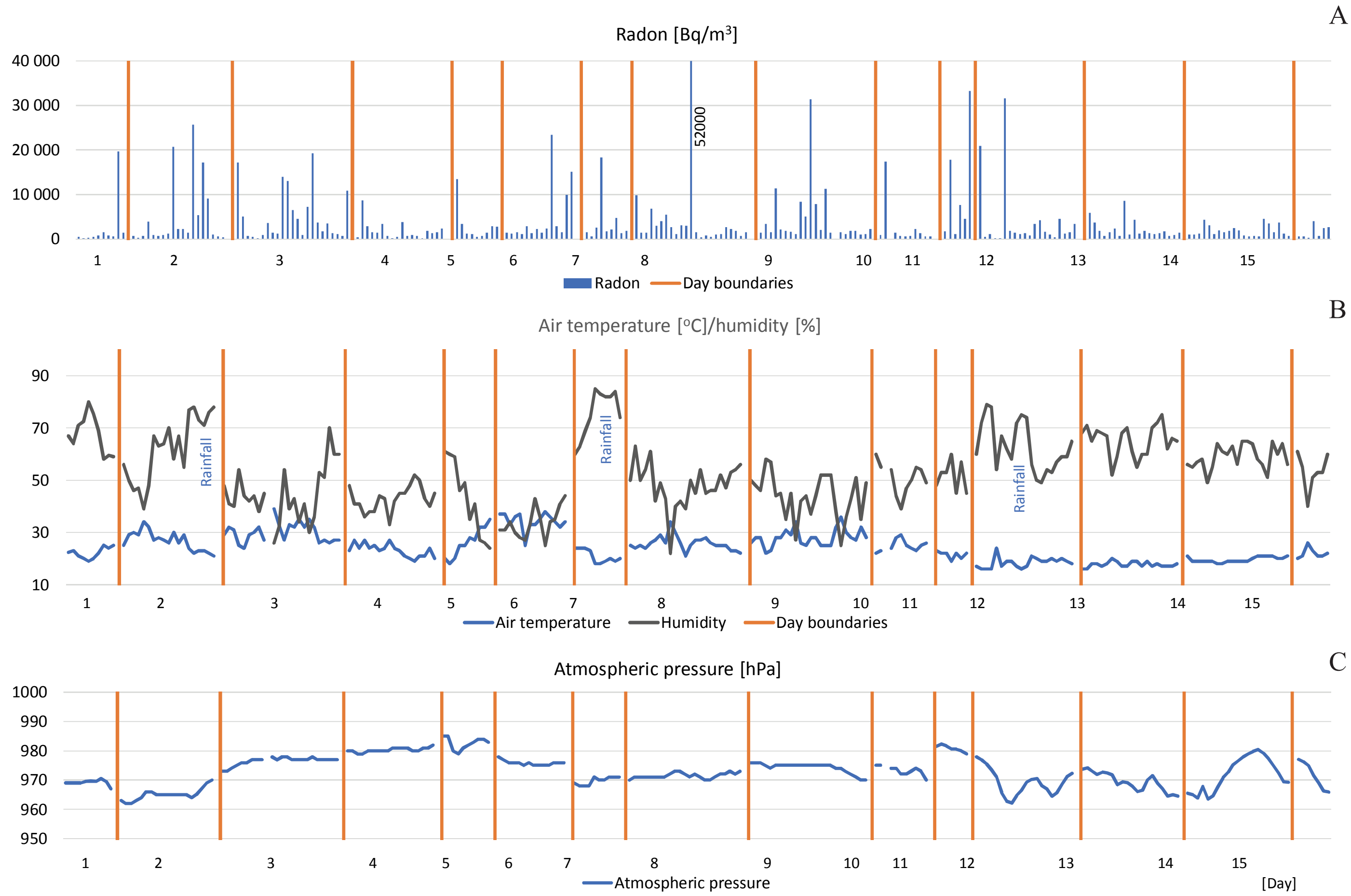

Fig. 3. Results of measurements: A – radon, B – air temperature and humidity, C – atmospheric pressure, during the 15 days of fieldwork

the weather conditions impact were carried out during 15 days, which comprised 240 measurement points determined along the Cergowa Mountain–Krosno profile with a step of 100 m. At each analysis of the soil air an independent measurement of atmospheric conditions was made at a height of approximate 1.5 m above the ground surface, measurement time was about 1 minute. Results are presented in three graphs in Figure 3.

The analysis of the above graphs did not show the influence of weather conditions on radon measurements in the soil air in the area of the Carpathians. The recorded anomalies reached even as much as 52 000 Bq/m^3, when the overall background did not exceed 1000 Bq/m^3. The humidity was checked due to the description of its impact on measurements in other publications and because of the gas concentration growth after the rainfall during stationary measurements. The fieldwork does not allow to repeat the entire measurement series, hence the data were analysed on the basis of results correlation and of statistical analysis. Throughout the whole measurement series the weather conditions were variable, in particular the air humidity and accompanying precipitation. On day two, seven and twelve, when there were rainfalls, monitored average variables were not higher than those collected in other days (Table 2). The results featured a lack of average radon correlation with the average air humidity (Figure 4), which proves a little influence of the monitored variables on the recorded values.

Table 2. Average radon concentration for every day of measurements

	Day	Average radon concentration [Bq/m^3]
High humidity	1	2620
Precipitation	2	5011
Medium humidity	3	5278
Medium humidity	4	1820
Medium humidity	5	2816
Low humidity	6	4896
Precipitation	7	3991
Medium humidity	8	4687
Medium humidity	9	4297
Medium humidity	10	2644
Medium humidity	11	9546
Precipitation	12	4104
High humidity	13	2171
High humidity	14	1873
Medium humidity	15	1596

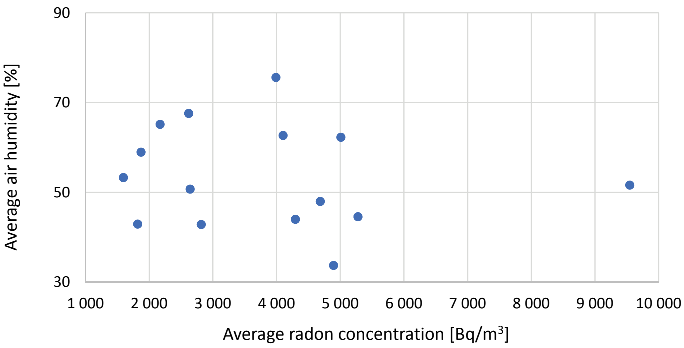

Fig. 4. Correlation graph for average daily air humidity and average daily radon concentration measured during fifteen days of research

Natural gamma – ray radiation of rocks nearby measurement points was tested to evaluate influence of potassium, uranium and thorium for radon concentration (Figure 5). In this case most important was comparison of uranium with radon which are in the same chain decay.

The natural radiation tests were carried out in the field on the Cergowa Mountain–Jasionka profile (Figure 5). It is a part of Cergowa Mountain–Krosno profile which was analysed in case of weather condition. This part of profile was chosen for research because of occurrence two different bedrock type which are below locations of radon measurements. Cergowa block is built of Cergowa Sandstones. The southern part of the mountain is composed of lower Krosno strata with sandstone-shale and of chaotic complexes. In the north direction from Cergowa, bedrock is built of the Oligocene-Miocene sediments (lower Krosno strata, shales) [11]. Point 34 in the figure 5 was neglected due the lack of measurement performance possibility on a steep edge of the mountain, it can be treated as a boundary between two different complexes.

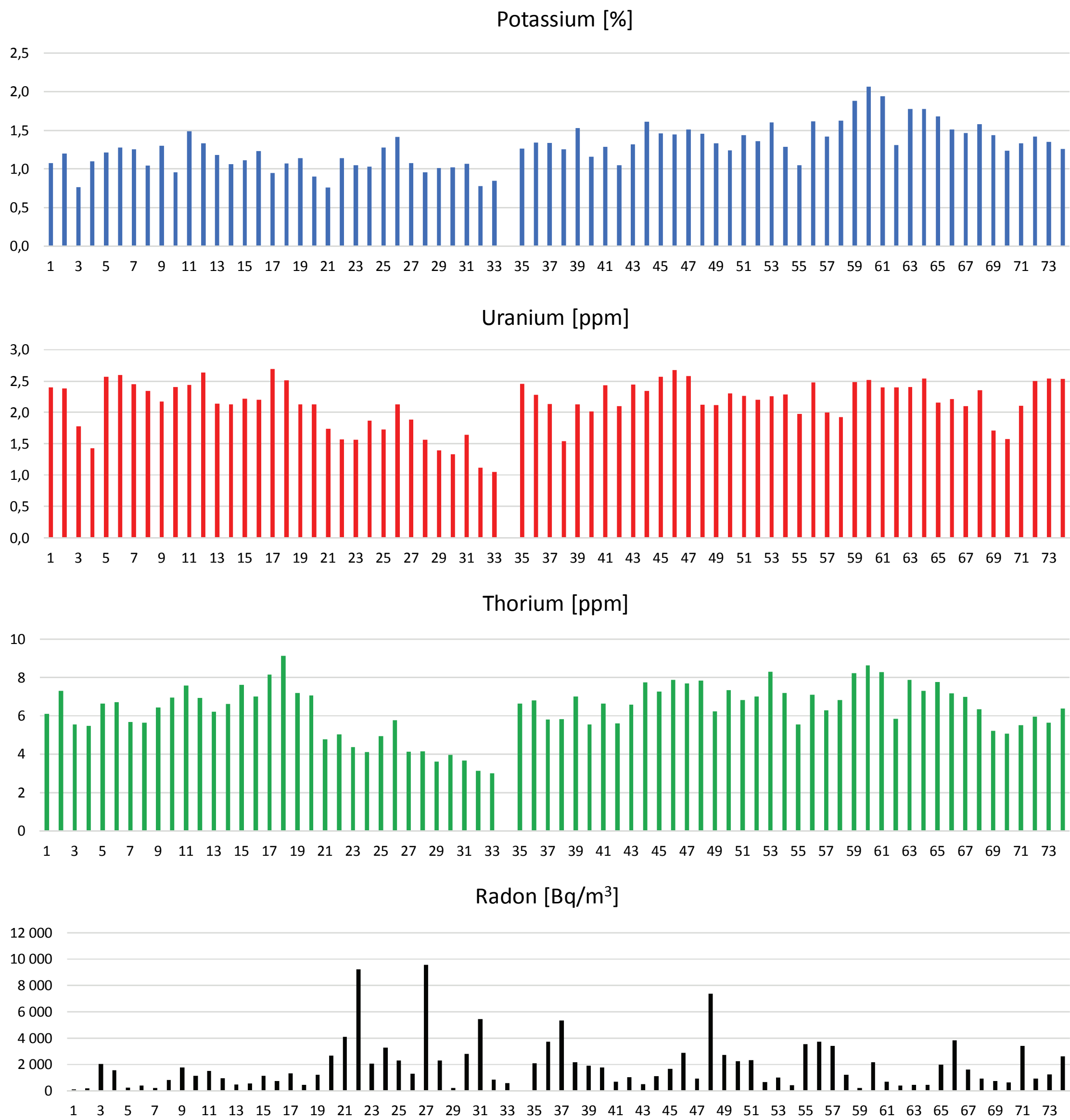

Fig. 5. Measured values for potassium, uranium, thorium, and radon along the Cergowa Mountain–Jasionka profile

Cergowa mountain and its southern part feature lower potassium values (significant majority of results below 1.4% for points 1÷33). Uranium and thorium concentrations above Cergowa Sandstones are also lower, in particular at points from 21 to 33 (less than 2 ppm). Higher concentrations of elements are situated in the area of Jasionka (points from 35 to 73), this region is covered with a thicker layer of soil profile, covering rocks built of the shales. Increased values for radon and anomalously three high values for three points in the Cergowa area (points from 19 to 33), proves a complicated deep structure of the mountain and its dissimilarity to the areas situated north and south. The analysis of rock radioactivity studies did not show a correlation with radon (Figure 6).

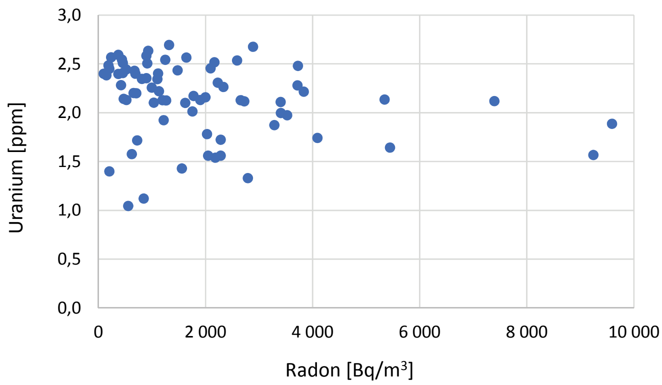

Fig. 6. Correlation graph of radon and uranium concentration measured in that same points in the field. Data collected in the Cergowa Mountain–Jasionka profile

Conclusions

Preliminary stationary studies in the soil profile allowed to assess the measurement errors of the method, to determine the measurement time (10 minutes), and to determine an important parameter – the time of detection chamber purging (variable). A series of measurements were performed immediately after the end of rainfall, which resulted in increased recorded values, what proved that such a weather conditions should be avoided.

The fieldwork, where anomalies featured extremely high scattering of values (from 30 Bq/m^3 to more than 50 000 Bq/m^3), did not show an influence of humidity, air temperature and atmospheric pressure for radon measurement results. Also the correlation of surrounding strata radioactivity with radon did not disturb the results. The lack of correlation with registered radon values was shown for uranium.

Collecting an accurate real radon values is not required for searching major fault zones. Instead, it is important that the measurement is precise and repeatable. A permanent methodology and retaining the same geometrical arrangement for each measurement point, including the measurement time, the sampling depth, and the probe length together with conductors are most important in the case of fieldwork. The other factors, like the air humidity, temperature, atmospheric pressure and radioactivity of surrounding strata can be assumed negligible in the case of carrying out studies to learn the arrangement of geological structures in the Carpathians. The structure of the analysed area provides exceptionally great anomalies of the recorded parameter, which facilitates the subsequent analysis of results and their interpretation. In previous publications, in which the impact of environmental conditions on results was described, the radon measurements were carried out seasonally and at much smaller data scattering. On the basis of carried out studies, the literature review and RAD7 specification it is recommended to carry out the measurements under similar weather conditions.

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