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Author: Maria Dziurowicz, Dariusz Malczewski, Jerzy Żaba

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^{222}Rn and ^{220}Rn concentrations in selected soils developed on the igneous rocks of the Kaczawa Mountains (Sudetes, Poland)

***Maria Dziurowicz*[1], *Dariusz Malczewski*[1]*and* *Jerzy Żaba*[1]**

This study presents the preliminary results of 222Rn (radon) and 220Rn (thoron) concentrations that were measured in the soils developed on igneous rocks including mafic (pillow lavas, basalts and melaphyre) and acid (keratophyre, rhyolite and rhyolitic tuff) rocks at ten locations in the Kaczawa Mountains (SW Poland). The measurements were carried out at sampling depths of 10, 40 and 80 cm using a RAD7 portable radon system. The highest concentrations of radon (222Rn) were mainly observed in the soils overlaying acid igneous rocks. The highest value of 11 kBq m-3 was obtained at a depth of 10 cm for soils overlaying melaphyre. At depths of 40 and 80 cm, the averages of 222Rn concentrations showed the same values of 3.6 kBq m-3 for all of the soils investigated. The highest concentrations of thoron (220Rn) were observed in soils overlaying acid igneous rocks, i.e. the value of 49 kBq m-3 at a depth of 40 cm for soils overlaying rhyolitic tuff. In the soils developed on basalts, the average concentrations of 222Rn increased with the sampling depth, whereas the average concentrations of thoron (220Rn) decreased with increasing sampling depth. Positive correlations were found between 232Th activity in the parent rocks and soil gas 220Rn concentrations at all of the sampling depths, whereas a positive correlation between soil gas 222Rn and the 238U activity concentration in the parent rocks was only found at a depth of 40 cm.

Key words*: Rn, radon concentrations, thoron concentrations, soil gas, in situ measurements. Sudetes.*

Introduction

Radon is an odourless, colourless, inert gas with isotopes of relatively short half-lives, two of which are naturally present in the environment with significance for practical purposes - radon 222Rn (T1/2 = 3.64 d) belongs to the 238U decay series as a product of the α-decay of 226Ra and thoron 220Rn (T1/2 = 55.6 s) belongs to the 232Th decay series as a product of the α-decay of 224Ra (Firestone, 1996). The applicability of both 222Rn (radon) and 220Rn (thoron) has become increasingly recognised as tracers in the field of environmental geosciences (Khattak et al., 2011), among others, in hydrology for investigations of groundwater migration processes and in earthquakes and volcanic activity as a possible precursor of these events (Cox et al., 1980; Gervino et al., 2012; Ramola et al., 1990). Some investigations have been done to measure 222Rn concentrations for soils developed on fault zones and uranium deposits (King, 1978; Solecki 1997; Malczewski and Żaba, 2007).

As a radioactive gas naturally emitted from the ground, radon has recently become an excellent tool in every aspect of geological research. The radon concentration values are correlated with the geological characteristics of the area studied. Radon measurements have usually been carried out in regions where high levels could have been expected (Malczewski and Żaba, 2007).

In the present study, the authors report the first results of 222Rn (radon) and 220Rn (thoron) concentrations measured at various depths in soils, mostly regosols and lithosols, developed on the igneous rocks at ten locations in the area of the Kaczawa Mountains in southwestern Poland (Fig. 1). These measurements presented here were taken in June 2014.

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[1] *Maria Dziurowicz, Dariusz Malczewski, Jerzy Żaba*, The Faculty of Earth Sciences, The University of Silesia, Będzińska 60, 41-200 Sosnowiec, Poland, maria.dziurowicz@us.edu.pl, dariusz.malczewski@us.edu.pl, jerzy.zaba@us.edu.pl

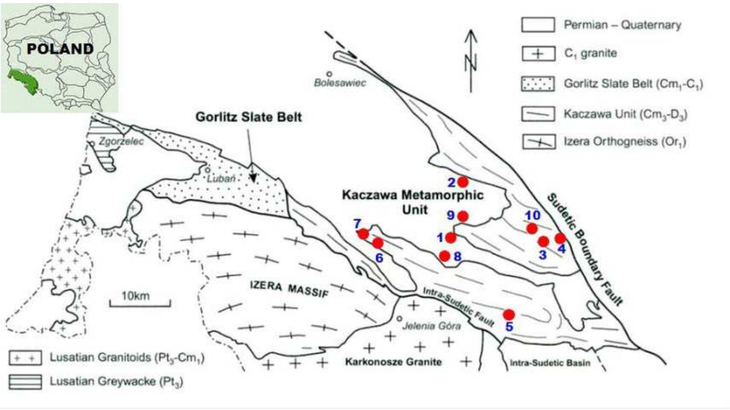

Fig. 1. Schematic map of the Kaczawa Unit in the West Sudetes showing in situ measurement locations 1-10. Age assignments: Pt, Proterozoic; Cm, Cambrian; Or, Ordovician; D, Devonian; C, Carboniferous; 1, early; 3, late. After: Aleksandrowski and Mazur, 2002.

The Kaczawa metamorphic complex (the Lower Unit of the Kaczawa Mountains) is the largest geological unit of the Sudetes and is composed of Paleozoic low-grade metamorphic rocks of sedimentary and volcanic origin. The age of these rocks dates from the Cambrian/Ordovician period through the Lower Carboniferous (Aleksandrowski and Mazur, 2002; Kryza et al., 1990). Tertiary tectonic activity terminated the development of the Miocene-aged basalts.

The main purpose of the study was to show the preliminary results of measurements of 222Rn (radon) and 220Rn (thoron) concentrations in soils developed on igneous rocks in the area of the Kaczawa Mountains as a function of the 238U and 232Th activity concentrations in the parent rocks. The relationships between the radon and thoron concentrations and meteorological factors such as temperature, atmospheric pressure and relative humidity were not performed here because the variability of meteorological parameters is complex and requires more experimental data.

Materials and Methods

In-situ measurements of ^{222}Rn and ^{220}Rn concentrations in the soils in the Kaczawa Mountains were carried out using a RAD7 portable radon system (Durridge Company, Inc.) working in sniff mode. The system contains a solid-state ion-implanted planar silicon detector and a built-in pump with a flow rate of 1 L min^{-1}. Inlet filters with a pore size of 1 μm block fine dust particles and radon daughters from entering the test chamber. The alpha spectrometry that is used by RAD7 system permits the signals of the ^{222}Rn and ^{220}Rn progeny to be spectrally separated as these have different alpha energies. The detector operates in a wide range of external relative humidity from 0 % to 95 % and internal humidity of 0 % to 10 % with a sensitivity of 4 Bq m^{-3} and an upper linear detection limit of 800 kBq m^{-3} (User Manual: Durridge Company Inc., 2000).

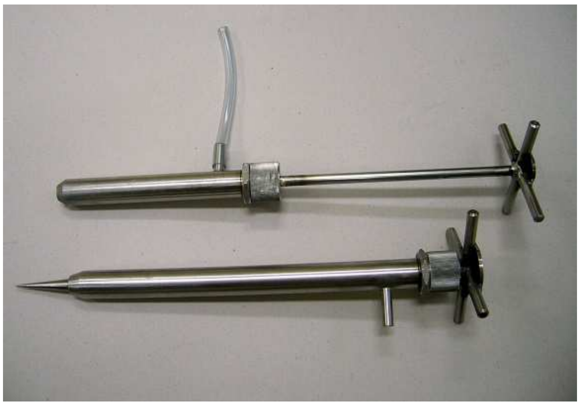

Fig. 2. 16 cm long soil gas probe.

The 16, 46 and 106 cm long soil gas probes that were used for the field work were made of stainless steel (Fig 2). These probes were inserted into the soil at depths of 10, 40 and 80 cm. After inserting the probe at the specified depth, the sampling outlet was connected to the inlet of the RAD7 via a small drying tube, which allowed the air to be removed from the soil and delivered directly to the detector chamber without dilution by outside air. In our measurements, the cycle time was 15 min and three cycles were performed. A single run at a specified depth was 45 min. The final result was the mean of these three measurement cycles.

The activity concentrations of primordial radionuclides (^{40}K, ^{232}Th and ^{238}U) in the igneous parent rocks of the Kaczawa Mountains were determined using an HPGe gamma-ray spectrometry system (Makowska, 2010). The system GX3020 (Canberra Industries) is based on coaxial HPGe Extended Range detector with a relative efficiency of 32 %, energy resolution of 0.86 keV at 122 keV and 1.76 keV at 1332 keV, and detector bias voltage of 4000 V. The rock samples, after crushing and then six-month collection, were measured in Marinelli 450 containers with the total duration of a single measurement of two days. For the efficiency calibration, determination of radionuclides and their activities the LabSOCS (Laboratory Sourceless Calibration Software) and Genie 2000 v.3 software packages were used. The activities of the particular radionuclides were calculated from the following gamma transitions (energy in keV): non series - ^{40}K (1460.8), thorium series - ^{208}Tl (510.8, 583.1, 860.6 and 2614.5), ^{212}Pb (238.6 and 300.0), ^{228}Ac (338.3, 911.6 and 969.1), uranium series - ^{214}Pb (241.9, 295.2 and 351.9) and ^{214}Bi (609.3, 1120.3 and 1764.5).

Results and Discussion

The soil gas ^{222}Rn and ^{220}Rn concentrations for all ten measurement locations are given in Table 1.

Tab. 1. Soil gas ^{222}Rn and ^{220}Rn concentrations at specified depths in soils developed on igneous rocks. Uncertainties are quoted as two standard deviations of counting.

No.	**Measurement Locations**	**Parent rock**		**Meteorological conditions** **Temperature (t) Relative humidity (RH)**	**Sampling depth** [cm]	**^{222}Rn (radon) concentration** [Bq m^{-3}]	**^{220}Rn (thoron) concentration** [Bq m^{-3}]
		Type	**Age**				
1	Wielislaw Organ N51^{o}02.091’ E015^{o}52.126’ h = 202 m	Rhyolite	Permian	t=24.6^{0}C RH= 23%	10	4262 ± 736	11563 ± 824
				t=26.9^{0}C RH=19%	40	6200 ± 1642	6020 ± 697
				t=24.9^{0}C RH=22%	80	9898 ± 2031	5593 ± 565
2	Wilcza Mountain N51^{o}06.328’ E015^{o}54.712’ h = 262 m	Basalt	Miocen	t=32.4^{0}C RH= 7%	10	517 ± 100	6916 ± 1018
				t=28.6^{0}C RH=23%	40	1198 ± 129	2303 ± 292
				t=33.6^{0}C RH=24%	80	1262 ± 147	1219 ± 124
3	Myślibórz Gorge N51^{o}00.904’ E016^{o}05.997’ h = 303 m	Pillow lava	Cambrian	t=22.1^{0}C RH= 14%	10	327 ± 48	660 ± 214
				t=24.6^{0}C RH=30%	40	430 ± 130	532 ± 176
				t=23.4^{0}C RH=31%	80	832 ± 120	473 ± 144
4	Myślibórz Small Organ N51^{o}01.304’ E016^{o}07.718’ h = 347 m	Basalt	Miocen	t=21.2^{0}C RH= 21%	10	1572 ± 158	2626 ± 270
				t=22.2^{0}C RH=24%	40	4873 ± 1204	1431 ± 172
				t=21.4^{0}C RH=25%	80	5297 ± 1281	1262 ± 214
5	Mysłów N50^{o}54.920’ E015^{o}58.443’ h = 493 m	Keratophyre	Ordovician	t=26.6^{0}C RH= 5 %	10	2658 ± 330	26866 ± 1256
				t=25.0^{0}C RH= 4%	40	6245 ± 440	20051 ± 923
				t=29.0^{0}C RH=22%	80	1861 ± 409	3928 ± 395
6	Wleński Gródek N51^{o}01.051’ E015^{o}39.752’ h = 342 m	Pillow lava	Cambrian	t=12.4^{0}C RH= 35%	10	1435 ± 303	1532 ± 360
				t=11.4^{0}C RH=37%	40	3354 ± 353	802 ± 162
				t=10.6^{0}C RH=24%	80	6085 ± 1017	3756 ± 320
7	Wleński Gródek N51^{o}00.771’ E015^{o}39.397’ h = 397 m	Basalt	Miocen	t=11.7^{0}C RH= 22%	10	903 ± 236	9433 ± 698
				t=19.4^{0}C RH= 4%	40	258 ± 96	3427 ± 350
				t=19.5^{0}C RH=29%	80	7136 ± 626	2908 ± 172
8	Lubiechowa N50^{o}59.842’ E015^{o}50.375’ h = 365 m	Melaphyre	Permian	t=17.3^{0}C RH= 4 %	10	11324 ± 1382	10068 ± 1128
				t=20.5^{0}C RH=26%	40	4489 ± 564	2554 ± 502
				t=18.2^{0}C RH=27%	80	1121 ± 484	1033 ± 144
9	Dynowice N51^{o}03.427’ E015^{o}51.675’ h = 181 m	Rhyolitic tuff	Permian	t=25.2^{0}C RH= 8%	10	1000 ± 255	17922 ± 1430
				t=27.3^{0}C RH=26%	40	2305 ± 194	48842 ± 4074
				t=26.1^{0}C RH=26%	80	1233 ± 207	12091 ± 629
10	Czartowska Rock N 51^{o}02.15’ E016^{o}01.687’ h = 442 m	Basalt	Miocen	t=18.3^{0}C RH= 8%	10	1161 ± 270	2656 ± 234
				t=21.6^{0}C RH=25%	40	6505 ± 953	3561 ± 578
				t=18.8^{0}C RH=26%	80	860 ± 277	1329 ± 225

At 10 cm

Soil gas ^{222}**Rn concentrations at a depth of 10 cm** (Tab. 1) varied from 327 Bq m^{-3} at point 3 (pillow lava, Myślibórz Gorge) to 11.3 kBq m^{-3} at point 8 (melaphyre, Lubiechowa) with an arithmetic mean of ca 2500 Bq m^{-3}. It is interesting to note that for soil developed on melaphyre, which is a volcanic rock similar to basalt in origin and appearance (palaeozoic basalt), the value of the ^{222}Rn concentration was exceptionally high in comparison with the other locations at the same depth. The arithmetic mean without the value for melaphyre equalled 1550 Bq m^{-3}, i.e. nearly two times less. However, in order for the authors to come to the appropriate conclusion, additional measurements at this location are required.

The soil gas ^{220}**Rn concentrations at a depth of 10 cm** (Tab. 1) varied from 660 Bq m^{-3} (Myśliborz Gorge, pillow lava) to 27 kBq m^{-3} (Mysłów, keratophyre) with an arithmetic mean of 9024 Bq m^{-3}.

At 40 cm

Soil gas ^{222}**Rn concentrations at a depth of 40 cm** (Tab. 1) varied from 258 Bq m^{-3} at point 7 (basalt, Wleński Gródek) to 6506 Bq m^{-3} at point 10 (basalt, Czartowska Rock) with an arithmetic mean of ca 3600 Bq m^{-3}. The highest concentrations (> 6000 Bq m^{-3}) were recorded at three locations, i.e. at points 1 (rhyolite, Wielisław Organ), at point 5 (keratophyre, Mysłów) and at point 10 (basalt, Czartowska Rock). Exceptionally low ^{222}Rn concentrations (< 500 Bq m^{-3}) were noted at points 3 (pillow lava, Myślibórz Gorge) and 7 (basalt, Wleński Gródek). The concentrations of ^{222}Rn measured at a depth of 50 cm in soil gas developed on granitic rocks between 133 Bq m^{-3} and 143 kBq m^{-3} were reported in Northern Malaysia (Almayahi et al., 2013).

Soil gas ^{220}**Rn concentrations at 40 cm** (Tab. 1) varied from 532 Bq m^{-3} (Myśliborz Gorge, pillow lava) to 49 kBq m^{-3} (Dynowice, rhyolitic tuff) with an arithmetic mean of 8952 Bq m^{-3}. The concentrations of ^{220}Rn at a depth of 50 cm that ranged from 35 to 403 Bq m^{-3} were observed in Malaysia (Almayahi et al., 2013).

At 80 cm

At **a depth of 80 cm** (Tab. 1), the highest soil gas ^{222}**Rn concentration** of 9899 Bq m^{-3} was found at location 1 (rhyolite, Wielisław Organ), and the lowest value of 833 Bq m^{-3} was measured at location 3 (pillow lava, Myślibórz Gorge) with an arithmetic mean of 3600 Bq m^{-3}, which is significantly lower than an average (32 kBq m^{-3}) reported for ^{222}Rn in a study of soils developed on metamorphic and sedimentary rocks at 80 cm in the Kaczawa Mountains (Wołkowicz, 2007). In the Kaczawa Mountains the average concentrations of ^{222}Rn at 10, 40 and 80 cm are definitely lower than those observed in similar soils in the neighbouring Karkonosze-Izera Block, i.e. 8, 78 and 224 kBq m^{-3}, respectively (Malczewski and Żaba, 2007). An average concentration of ^{222}Rn in soil gas developed on weathered granite of 100 kBq m^{-3} was reported in Guangdong Province, China (Wang et al., 2012).

The soil gas ^{220}**Rn concentrations at a depth of 80 cm** (Tab. 1) ranged from 473 Bq m^{-3} (Myśliborz Gorge, pillow lava) to 12 kBq m^{-3} (Dynowice, rhyolitic tuff), with an arithmetic mean of 3359 Bq m^{-3}. In the Kaczawa Mountains region the average concentrations of ^{220}Rn at 40 and 80 cm are lower than those observed in similar soils in the neighbouring Karkonosze-Izera Block, i.e., 12.7 and 16.3 kBq m^{-3}, respectively (Malczewski and Żaba, 2007). At 10 cm these concentrations were comparable - 9024 and 5450 Bq m^{-3}, respectively. A significantly higher average concentration of ^{220}Rn of 294 kBq m^{-3} at a depth of 80 cm was recorded in Guangdong Province, China (Wang et al., 2012). However, the authors did not explain the origin of such high concentrations of ^{220}Rn in the investigated soil gas.

Concentrations of ^{222}Rn in soils developed on basalts

In the Kaczawa Mountains measurements of ^{222}Rn concentrations in the soils developed on basalts were taken at four locations: 2 (Wilcza Mountain), 4 (Myślibórz Small Organ), 7 (Wleński Gródek) and 10 (Czartowska Rock). The data that were collected permitted the authors to draw preliminary conclusions regarding these soils at the specified depths. As can be seen in Fig. 3, the soil gas ^{222}Rn concentrations increase with increasing sampling depth with the average values of 1000, 3200 and 3600 Bq m^{-3} for depths of 10, 40 and 80 cm, respectively.

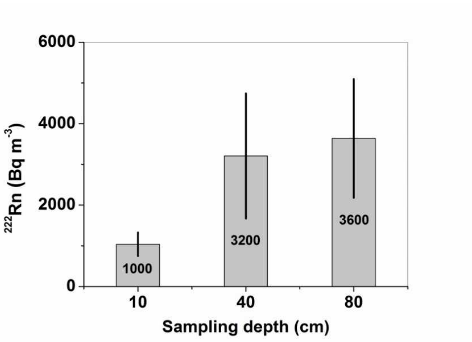

Fig. 3. Average soil gas 222Rn concentrations at points located on soils developed on basalts vs. sampling depth.

Concentrations of ^{220}Rn in soils developed on basalts

Measurement points located on soils developed on basalts were characterised by higher ^{220}Rn concentrations at 10 cm than at 40 and 80 cm. Fig. 4 shows a decrease in the average soil gas ^{220}Rn concentrations with increasing sampling depth. A similar effect of ^{220}Rn concentrations at points located on the slopes were observed in the Karkonosze-Izera Block (Malczewski and Żaba, 2007). More experimental data are needed to explore this effect.

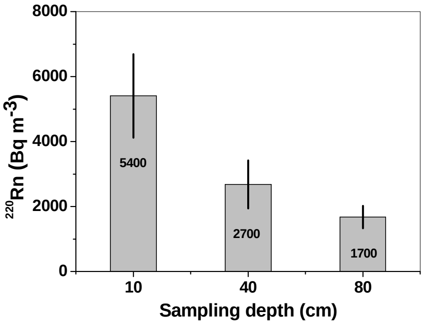

Fig. 4. Average soil gas ^{220}Rn concentrations at points located on soils developed on basalts vs. sampling depth.

Comparing Fig. 3 and 4, it is visible that the soil gas ^{222}Rn concentrations increased with increasing sampling depth, whereas the soil gas ^{220}Rn concentrations decreased with increasing sampling depth in soils developed on basalts. There are a few possible ways to interpret the different relationships for thoron and radon. Most of the thoron decay processes occur in soil with a relatively high concentration of thorium and therefore the ^{220}Rn concentration is expected to be high. The ^{220}Rn exhalation tends to decrease with an increase in depth, particularly when measured near the surface of basalts, which is why lower ^{220}Rn concentration values were measured at 80 cm (Fig. 4). Unlike ^{220}Rn, ^{222}Rn has a long half-life and it can accumulate in significantly higher concentrations; so, slight variations were observed in the ^{222}Rn concentrations at depths of 40 and 80 cm (Fig. 3).

^{222}Rn and ^{238}U correlations

As can be seen in Fig. 5a, the soil gas ^{222}Rn concentrations at a depth of 10 cm and parent rock ^{238}U activities were uncorrelated (r = - 0.02). When excluding point 8 (melaphyre), the correlation coefficient becomes significantly positive, r = 0.40 (Fig. 5b).

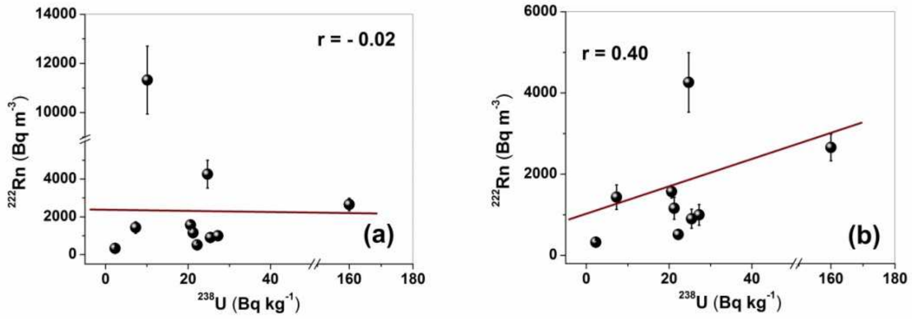

*Fig. 5. Soil gas ^{222}Rn concentration at a depth of 10 cm vs. ^{238}U activity: **(a)** for all of the measurement locations and **(b)** without point 8 (melaphyre). The solid lines represent linear fits, r – correlation coefficient.*

At a depth of 40 cm, for all of the measurement locations, the correlation between ^{222}Rn concentrations and ^{238}U activity in the parent rocks was 0.41 (Fig. 6).

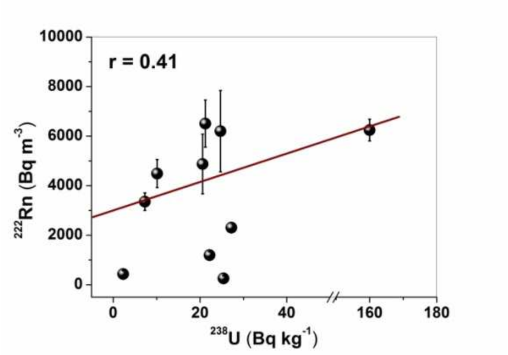

Fig. 6. Soil gas ^{222}Rn concentration at a depth of 40 cm vs. ^{238}U activity. The solid line represents a linear fit, r – correlation coefficient.

At a depth of 80 cm, the correlation ^{222}Rn – ^{238}U was weak and negative, r = - 0.13 (Fig. 7).

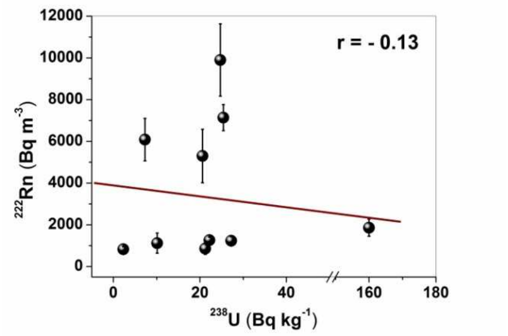

Fig. 7. Soil gas ^{222}Rn concentration at a depth of 80 cm vs. ^{238}U activity. The solid line represents a linear fit, r – correlation coefficient.

^{220}Rn and ^{232}Th correlations

As can be seen in Fig. 8, a strong correlation was observed (r = 0.93) between the ^{232}Th activity in the parent rocks and ^{220}Rn soil gas concentrations at a depth of 10 cm.

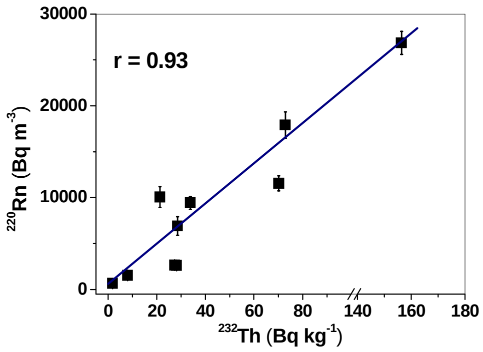

Fig. 8. Soil gas ^{220}Rn concentration at 10 cm vs. ^{232}Th activity. The solid line represents a linear fit, r – correlation coefficient.

At a depth of 40 cm, for all of the measurement locations, the correlation coefficient was 0.56 (Fig. 9a). When point 9 (rhyolitic tuff) was excluded the correlation coefficient increases to 0.96 (Fig. 9b).

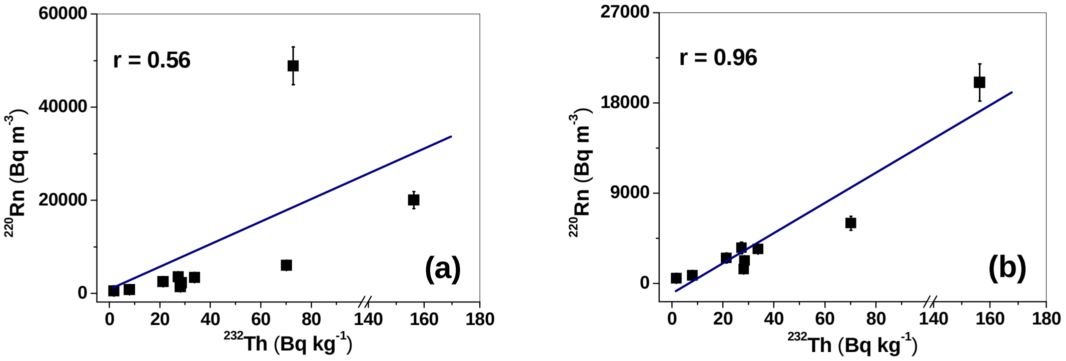

*Fig. 9. Soil gas ^{220}Rn concentration at 40 cm vs. ^{232}Th activity for all of the measurement locations **(a)** and **(b)** without point 9 (rhyolitic tuff). The solid lines represent linear fits, r – correlation coefficient.*

At 80 cm the correlation between the ^{232}Th activity in the parent rocks and soil gas ^{220}Rn concentrations was lower than for those at 10 and 40 cm but it was still significant (r = 0.45, Fig. 10). Without the rhyolitic tuff, the correlation was slightly higher (r = 0.56).

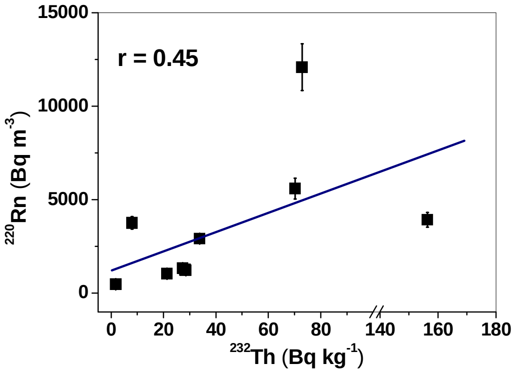

Fig. 10. Soil gas ^{220}Rn concentration at 80 cm vs. ^{232}Th activity. The solid line represents a linear fit, r – correlation coefficient.

^{220}Rn and ^{222}Rn correlations

The average concentrations of ^{220}Rn and ^{222}Rn for all of the profiles were essentially uncorrelated (r = - 0.07, Fig. 11a). However, excluding the concentrations in soils developed on keratophyre and rhyolitic tuff, the correlation coefficient was 0,68 (Fig. 11b). This probably results from the predominance of ^{220}Rn over ^{222}Rn at locations 5 and 9 as an effect of the high activity concentrations of ^{232}Th in the parent rocks.

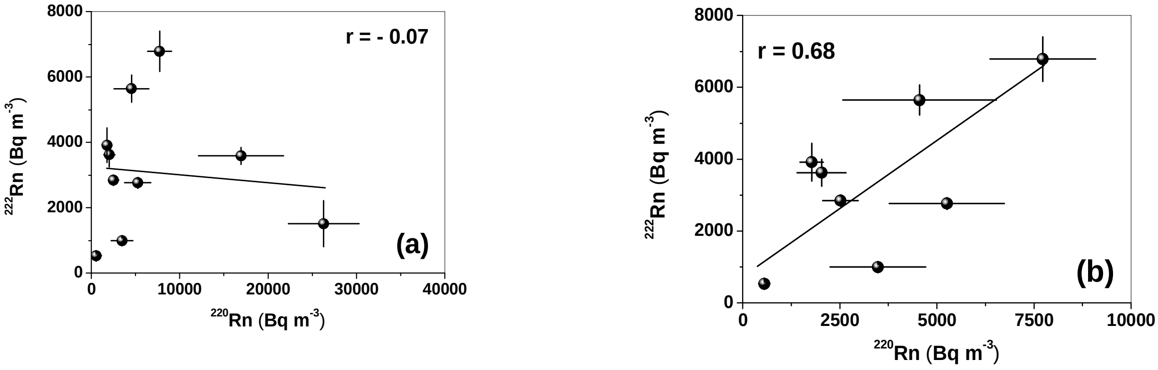

*Fig. 11. Correlation of ^{220}Rn and ^{222}Rn concentrations averaged for all of the depths **(a)** and **(b)** without points 5 (keratophyre) and 9 (rhyolitic tuff). The solid lines represent linear fits, r – correlation coefficient.*

Conclusions

Our preliminary study showed that a positive correlation was found between soil gas 222Rn concentrations and the 238U activity concentration in the parent rocks at a depth of 40 cm, whereas at sampling depths of 10 and 80 cm, these concentrations were uncorrelated. More experimental data are needed to explain this effect. Positive correlations between 232Th activity in the parent rocks and soil gas 220Rn concentrations were found at all of the sampling depths of 10, 40 and 80 cm. The average concentrations of 220Rn and 222Rn were correlated excluding the points that were located on soils that had developed on keratophyre and rhyolitic tuffs.

Measurements of the 238U and 232Th activity concentrations in soils compared to the 222Rn and 220Rn concentrations developed on the rocks of the Kaczawa Mountains are planned as the work presented here is still in progress.

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