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Temporal and spatial changes of radon concentration in borehole water (Little Carpathians Mts., Slovakia)

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Abstract. The ^{222}Rn activity concentration in ground water from four boreholes was investigated from January 2006 to June 2008. The boreholes are situated in the region of the Astronomical and Geophysical Observatory in Modra-Piesok (Little Carpathians Mts., 40 km NW from Bratislava, Slovakia). Three boreholes have been drilled in Lower Triassic quartzite. Another borehole has been drilled in granodiorite of the Modra massif in which the quartzite is folded. Temporal and spatial differences in radon concentration were observed. Significant short-term variations were noticed in all boreholes. Precipitation caused the changes of water level and strongly affected the values of ^{222}Rn activity concentration in less deep boreholes. The measured activities in boreholes ranged approximately over 1–240 kBq/m^3.

1 Introduction

Radon is important in the context of potable groundwater due to its high solubility in water. The greatest problems with waterborne radon occur in homes that are located in areas with high levels of radon in the groundwater, which are served by an individual well or small community water-supply system. Many private supplies have little or no treatment, minimal natural aeration, and a short time interval between pumping and consumption; therefore radon will not be dissipated (Clapham and Horan, 1996; Frengstad et al., 2003).

In Slovakia the natural radioactivity in water was investigated in 1990 by the State Geological Institute of Dionyz Stur within the scope of a project entitled "The investigations of geological factors of environment". Among other radionuclides, the ^{222}Rn content in water was determined. The median value for underground water in Slovakia was 9.75 kBq/m^3 (Daniel et al., 1996). However, the temporal variations of radon activity concentration were not studied.

WHO (2009) guidelines for drinking-water quality recommend a treatment of the water source, if the radon concentration exceeds a limit of 100 kBq/m^3. After the decree by the Slovak Ministry of Health 528/2007, the action level for radon activity concentration in public water supplies in the Slovak republic is 100 kBq/m^3 and the highest permissible limit is 300 kBq/m^3. The radon content in water sources is recommended to be examined once a year. Since the radon activity concentration in water is not stable during the year, the radon content determined in a single sample may not represent the true average value.

The aim of our research was to evaluate the temporal variations of radon activity concentration in borehole water in relation to the water level changes, precipitation amount and height of snow cover during a year. The effect of a different lithology on radon activity concentration was compared. The study site is located near a mountain holiday resort where cottages and houses have private wells. These wells are drilled in the same rock environments as the investigated boreholes; therefore the regular long-term water sampling in this project helps to elucidate the radon risk from drinking water to the population in the whole area. In this paper the results of the regular ^{222}Rn activity concentration monitoring in well water over a period of 2.5 years are presented.

2 Sampling site

The study site is located within the Little Carpathians Mts., near Modra town, at the Astronomical and Geophysical Observatory of Comenius University (AGO), approximately

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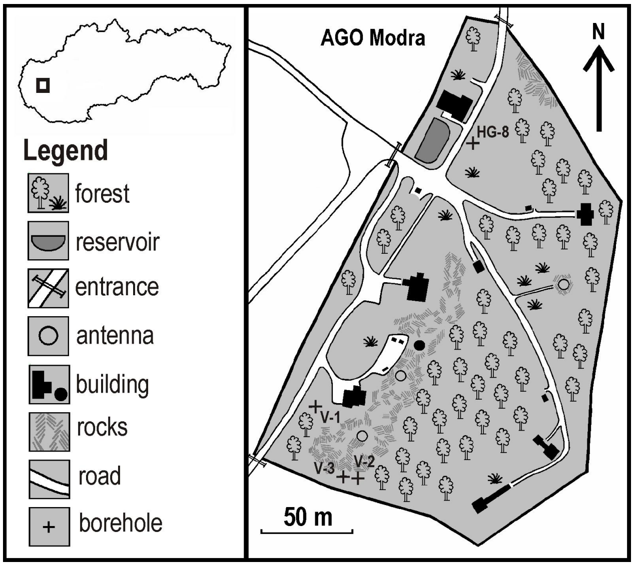

Fig. 1. Map of a sampling site with location of boreholes.

40 km NE from Bratislava. The geological surrounding of the AGO consists of the Modra granodiorite massif of Variscian age, of which the Lower Triassic quartzite is folded (Cambel and Vilinovič, 1997). Quartzite forms noticeable cliffs near the AGO and it is strongly cracked. The radon investigation in borehole air and water at AGO Modra started in August 2003 (Smetanová et al., 2006, 2008).

The water samples for the ^{222}Rn activity concentration analysis were collected from four boreholes located in two different lithologies (Fig. 1). The boreholes V-1 (10 m), V-2 (40 m) and V-3 (10 m) were drilled in quartzite. The inner diameter of these boreholes is 80 mm. They are reinforced with a PVC pipe with 10% perforation along their whole length.

The borehole HG-8 (50 m) was drilled to be used as a water source for the AGO. This borehole is 0.32 m in diameter. The borehole is reinforced by a steel pipe along its whole length. The pipe perforation ranged from 25 to 50 m. A water pump is placed at a depth of 38 m. The pumping of water from the well is irregular and depends on the water consumption in the AGO, which is approximately 1 m^3 per day. From the geological point of view this borehole is situated in two types of rock environment. Above the depth of 6 m below the surface the geology around the well consists of a stony detritus and soil mixture. The majority of this borehole is drilled in the hydrothermally altered granitoide of the Modra massif, and occasionally into pegmatites (Paňáková, 1985).

The whole area of the AGO Modra is located on a hill sloping to the north. The altitudes of boreholes are depicted in Table 1.

Table 1. Altitude of boreholes at AGO Modra.

Borehole	V-1	V-2	V-3	HG-8
Altitude [m a.s.l.]	531.48	537.46	537.77	512.75

3 Experimental methods

The water sampling was done regularly three times a week from January 2006 to June 2008. The samples were collected using a plastic bottle with a volume of 200 ml. After water collection each bottle was tightly sealed, the date and time of the sample collection was noted and the bottles were immediately transported to the laboratory. Simultaneously the water level state below the surface was measured in every borehole. The precipitation amount, the height of snow cover and atmospheric temperature have been regularly monitored at the AGO and kindly made available for our investigation.

The ^{222}Rn activity concentration in water was determined in the laboratory at the Department of Nuclear Physics and Biophysics, Faculty of Mathematics, Physics and Informatics of Comenius University in Bratislava. Measurement was based on the flushing of the water sample with non-active gas. Radon included in a 7 ml sample was transferred into a 125 ml scintillation cell of Lucas type (Lucas, 1957). After the radioactive equilibrium of radon with its daughters-products was achieved, the radon counts were measured. The total cumulative counts for every 200 s interval were recorded and counting continued up to 2000 s. The background of the detector is 0.3 cpm, the efficiency is 2.1 cps per one ^{222}Rn decay. The ^{222}Rn activity concentration in water was calculated using the equation

$$A\left(^{222}\mathrm{Rn}\right)\left[Bq/\mathrm{m}^3\right] = \frac{S-B}{K\ V}\,\frac{\lambda t\exp(\lambda\ T)}{1-\exp(-\lambda t)}, \qquad (1)$$

where S is sample counts per counting interval [s^{-1}], B is background counts per counting interval [s^{-1}], V is sample volume [m^3], K is detection efficiency, λ is ^{222}Rn decay constant [s^{-1}], T is time delay between sample collection and the beginning of counting measurement [s], t is the counting time [s].

4 Results and discussion

4.1 Short-term variation of ^{222}Rn activity concentration

The regular long-term water sampling showed significant non-periodic variation of ^{222}Rn activity concentration in all investigated boreholes, with a duration of 4–44 days. Rock bodies of both lithologies are strongly cracked. The significant difference in radon concentration and position of water level between V-2 and V-3 boreholes drilled only 8 m apart

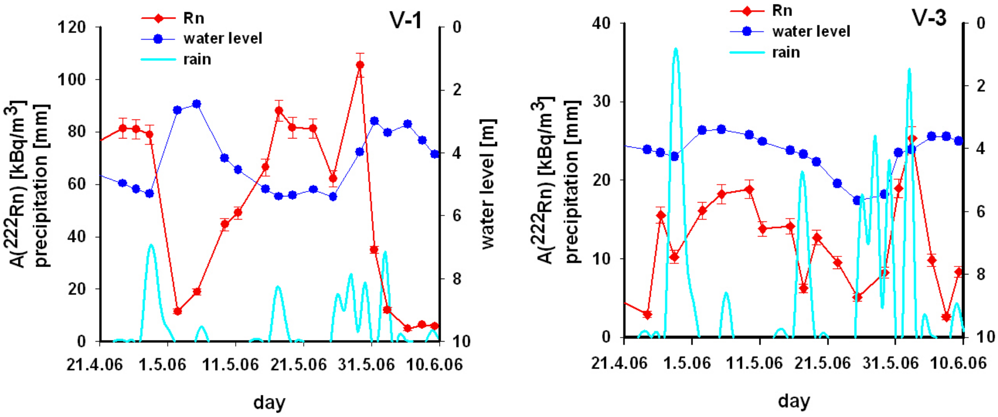

Fig. 2. Variation of radon activity concentration with rainfall in V-1 and V-3 boreholes. Vertical bars represent the standard deviation of the radon activity concentration determination.

in the same rock body demonstrates the different degree of crack connection. The water level in V-2 was situated approximately 23 m lower in comparison with V-3.

Large differences in radon activity concentration amongst boreholes were found. In V-3 radon activity concentration in water reached up to 30 kBq/m^3 only. The highest values measured in V-2 were comparable with the HG-8 borehole; they reached 250 kBq/m^3, while in V-1 maximum values were in order of 140 kBq/m^3. However, the elevated radon concentrations in V-1 and V-2 boreholes were found sporadically in comparison with HG-8, in which radon concentration ranged 150–250 kBq/m^3 with the exception of the winter months. Underlying granitoides are probably a radon source. Radon is transported into boreholes V-1 and V-2 drilled in quartzite by the fracture network. Low radon activity concentration in V-3 suggests that this borehole is not connected by fractures with the granodiorite body. Borehole water is surface water exclusively supplied by rainfall.

The precipitation effect on radon activity concentration in borehole water and the state of the water level was confirmed in V-1 and V-3 boreholes only. After a significant rainfall event (>20 mm/day), an increase in the water level up to 3 m in comparison with the previous state was noticed. Infiltrated water affected each borehole in a different way. In V-1 the precipitation was followed by an immediate steep decrease of radon activity concentration. Usually 2–10 days after the rainfall event the radon concentration started to increase until it achieved the values comparable to those before the rain. Contrarily, in the V-3 borehole radon concentration change due to rainfall was not unambiguously observed. When the soil was saturated with water, the delay between rainfall episode and radon increase was within a two day period, in the case of dry soil this period was 2–7 days (Fig. 2).

Radon activity concentration in water collected from the V-1 borehole decreased with increasing water level state (R=−0.64). When the water level in V-1 decreases to below 5 m under the surface, the decrease of radon activity concentration was observed (R=0.75). In the V-3 borehole radon concentration increased with an increase in the water level (R=0.81).

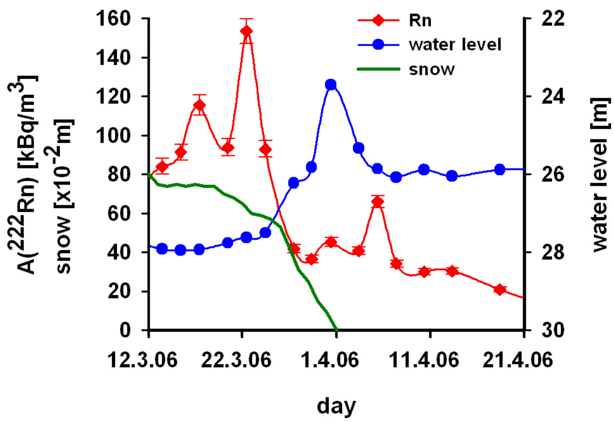

Fig. 3. Radon activity concentration decrease in V-2 borehole after the snow melting. Vertical bars represent the standard deviation of the radon activity concentration determination.

Boreholes V-2 and HG-8 were free from the rainfall influence. The water level was situated deep enough to be affected by infiltrated rainfall water. On the other hand, snow melting was accompanied by a noticeable increase of water level and the considerable decrease of radon concentration in water collected from both boreholes. This phenomenon was clearly observable in V-2 at the end of March 2006, when the thick snow layer of up to 1 m disappeared in 10 days. It resulted in the water level increasing by up to 5 m (Fig. 3). The snow cover in 2007 was poor and in 2008 was much thinner in comparison with 2006; however the snow melting was accompanied with a water level rise. Under this condition the change of radon concentration was not so clear. The effect of snow melting was observed in boreholes V-1 and V-3 for the whole period of the investigation. Water level increased up to 5 m and radon activity concentration in water decreased in both boreholes.

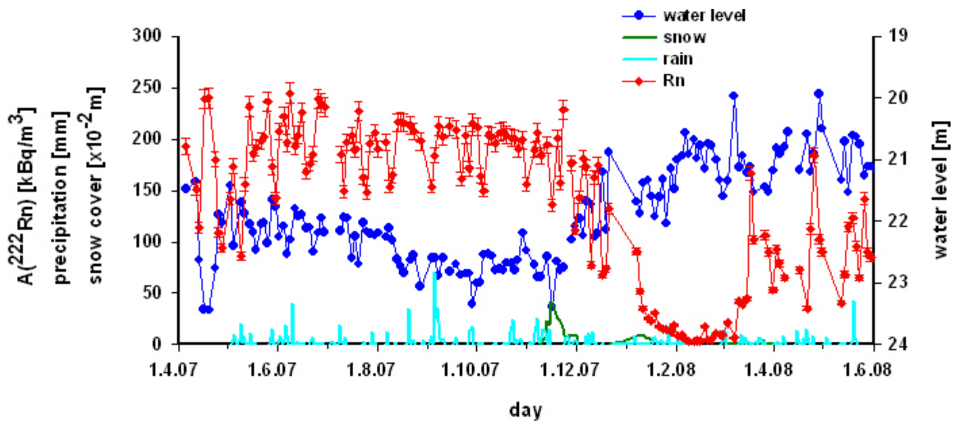

Fig. 4. Seasonal change of radon activity concentration and water level in HG-8 borehole. Vertical bars represent the standard deviation of the radon activity concentration determination.

4.2 Seasonal variation of ^{222}Rn activity concentration

Seasonal variation of ^{222}Rn activity concentration was found in the HG-8 borehole only, with the minimum in winter months. After the snow melt in November 2007 a rise of water level up to 2 m was noticed. At the same time the radon activity concentration in borehole water decreased to approximately 1 kBq/m^3, probably as the result of a dilution effect by a large influx of inactive water into the borehole (Fig. 4).

In the V-2 borehole the seasonal variation of radon activity concentration was difficult to evaluate. The seasonal change of water level occurred with maximum after the snow melting at the end of winter, but the changes of radon activity concentration were not the same for both years of investigation. Snow melting in autumn 2007 was not followed by the similar significant radon concentration decrease as in HG-8.

Due to the water deficiency in summer the seasonal change of radon activity concentration in water was impossible to observe in less deep V-1 and V-3 boreholes. The boreholes are drilled on a slope of a hill, as the groundwater drains away to the valley, the water level in boreholes decreased under the borehole base until intense precipitation or snow melting occurred.

4.3 Radon risk from water consumption

The HG-8 borehole is solely used as a water source. Water from this borehole is automatically pumped into a tank with a volume of 1 m^3, immediately after the water level in the tank decreases below a minimum level. Water stays in the tank for approximately one day, because of water consumption. Therefore radon activity concentration in tap water at the observatory is very similar to that measured in the water sample collected directly from the borehole, the difference in radon activity concentration is about 15%.

The action level of 100 kBq/m^3 for radon activity concentration in public water supply was exceeded in HG-8 borehole. The elevated radon activity concentration in this borehole ranged from 100 to 250 kBq/m^3 and was measured from spring to autumn months. However, the highest permissible limit was not reached.

The long term regular water monitoring showed that the collection of a single representative sample is insufficient for the purpose of radon risk determination. Radon activity concentration in water of both rock types seems to be the lowest after the snow melting. For that reason the representative water sample should be collected in summer months, but the collection of several samples during the year is better. In the case of boreholes with the water level situated near to the surface the water sampling should not be performed after an intensive rainfall event, when the results could be underestimated.

5 Conclusions

Radon activity concentration and water level state in boreholes at AGO Modra were unstable. Boreholes with water levels near the surface were strongly affected by precipitation and snow melting. Rainfall impact was not observed in deep boreholes. Seasonal change in ^{222}Rn activity concentration was confirmed in the HG-8 borehole exclusively, with a minimum after the snow melt. In the summer months water level in the V-1 and V-3 boreholes decreased so that groundwater was below the base level of the boreholes, probably as a result of evapotranspiration and groundwater flow down to the valley below.

The effect of lithology on radon concentration in water was observed. Highest radon activity concentrations were found in HG-8 borehole drilled in granodiorite. However, the highest measured radon activity concentration in V-2 and HG-8 reached similar values. Large variability of radon activity concentration was observed and for that reason the collection of one sample for radon risk determination seems to be insufficient.

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