4. Monitoring the Volumetric Activity of Radon in the Geological Environment of the Slovak Republic

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Abstract: Natural radionuclides and products of their radioactive decay are omnipresent in all the compartments of our environment. The radon volumetric activity in ground air, waters and above tectonics has been monitored in the Slovak Republic for several decades under umbrella of the geological task of the State Geological Institute of Dionýz Štúr "Partial Monitoring System of Geological Factors, Subsystem 05 -Monitoring of Radon Volumetric Activity". The several ten years of observations enable to distinguish long-term and short-term variations of radon content in selected sites and environs across Slovakia. The seasonal variations of the radon volumetric activity doesn't depend solely upon humidity and gas permeability of soils and rocks at the given site, but also on geological setting and lithology. The same meteorological conditions, but different geological environ result in different nature of these variations. This contribution presents the results of the radon activity monitoring over a period of 2002-2016, which has confirmed wavy or sinusoidal dependence in the radon bulk activity variations.

Keywords: radon; radioactive decay; radon volumetric activity; radon variations

4.1 Introduction

The human population is permanently exposed to the effects of different types of radiation. However, the issue of natural but also artificial (induced) radioactivity has been virtually "taboo" for decades. Uranium was a strategic raw material for the nuclear (arms, energy) industry, and the results of survey and scientific research in this area (particularly during the so-called "Cold War") were strictly classified. Health risks and impacts on the population have largely been degraded, and it has often been argued that radon and its nuclear decay products directly threaten miners in uranium mines.

Concerns and attention of the public have long been focused on artificial sources of radiation (nuclear weapons, nuclear power, health, etc.), with the most of the population not even knowing that the most significant irradiation (beyond the periods after nuclear experiments, accidents, etc.) is generated by natural sources (Fig. 4.1).

The impact of exposure to radon abroad is dealt with by several reputable institutions (e.g. UNSCEAR – *United Nations Scientific Committee on the Effects of Atomic Radiation*, ICRP – *International Commission on Radiological Protection*), but also other organizations and scientific workplaces. Their research has shown that natural sources of radioactive radiation make up nearly three quarters (73 %) of the total radiation burden of the population.

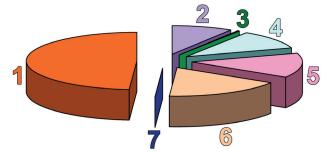


Fig. 4.1 Radiation burden of the population (web site: National RadiationProtectionInstitute, www.suro.cz)
1: radon (48.9 %), 2: natural radionuclides in the human body (8.9 %), 3: Chernobyl fallout (0.3 %), 4: medical exposure (10.9 %), cosmic (13.9 %), 6: gamma radiation from Earth (17.0 %), 7: others (0.13 %) – of which nuclear power plant output equals to 0.04 %

The most important source of natural radiation are radon (²²²Rn) and radionuclides (²¹⁸Po, ²¹⁴Po, ²¹⁰Tl, ²¹⁰Po,...) arising within its nuclear decomposition. These are adsorbed in the human airway where lung tissue cells are irradiated at the contact, which can ultimately lead to the development of malignant carcinoma.

Various studies have shown that the likelihood of lung cancer increases with the increasing concentration of radon and its decay products, but also with the duration of exposure. The fact that radon is the second most important cause of lung cancer after smoking is also recognized by the World Health Organization which classified it in 2009 in the class 1A carcinogen.

The results of newer epidemiological studies have shown that exposure to radon results in other forms of health damage such as vascular and digestive disorders. The likelihood of health damage is apparently significantly higher than originally assumed.

This has led the European Commission (EC), the International Commission on Radiological Protection (ICRP) and the World Health Organization (WHO) to reconsider health risk from radon exposure.

The European Commission has also defined new requirements to increase the protection of the population from the adverse effects of ionizing radiation – the elaboration of legislation aimed at protecting the population and the introduction of radon programmes in which population awareness is among the priorities.

In Slovakia, since the early 1990s, much more attention has been devoted to natural radioactivity and the risk

of exposure to radon from the geological environment when maps of natural rock radioactivity (concentration of potassium – 40 K, equivalent uranium – eU, equivalent thorium – eTh, total natural radioactivity – eUt, dose gamma radiation power – Da), radon risk forecast maps volume activities of radium 226 Ra and radon 222 Rn), radon 222 Rn in the ground air, and natural radioactivity in groundwater and surface water maps (U_{nat} = 234 U, 235 U and 238 U isotope concentration). Monitoring of short-term (seasonal) and long-term changes in radon content in the geological environment had not been the focus of solved tasks and no more attention had been paid to variations in radon volumetric activity (RVA; also term radon bulk activity, RBA, is used).

Since the project "Partial Monitoring System of Geological Factors (PMS GF)" has been solved, the solvers of the subsystem "Monitoring of Radon Volumetric Activity in the Geological Environment of the Slovak Republic" started to systematically address the issue in the whole Slovak Republic.

4.2 Characteristic of the topic in question

A whole range of natural radionuclides and their nuclear decay products are permanently present in all environmental compartments, i. e. in the rocks, in the waters and in the air. The radon (isotope ²²²Rn – source of alpha radiation with a half – life of 3.825 days) is an inert natural radioactive gas generated by the spontaneous disintegration of ²²⁶Ra (alpha and gamma radiation source) in the disintegrating range of uranium ²³⁸U and belongs to the so-called rare gases. It has a higher density than air, is very well soluble in water (up to 51% of its volume), it is colourless, tasteless and odourless.

The main source of natural radon is the geological environment, i.e. some minerals and rocks, as well as groundwater flowing through rocks with increased uranium content. Due to the half – life of maternal elements

(238 U = 4.47 x 109 years, 226 Ra = 1,600 years), the geological environment ensures its continuous supply.

The radon is relatively easy to penetrate through the rock environment and spreads through by diffusion or a convective flow. The diffusion is influenced by physical properties of the environment – especially porosity and moisture. The transport of the radon by convection is higher in order and is caused by the changes in the physical conditions of the environment (temperature and pressure gradients). The most significant manifestation of convection is over the tectonically disturbed zones, which provide good communication paths for gases.

In the free environment, radon is rapidly diluted with atmospheric air, but it can accumulate in high, health-harmful concentrations in confined spaces.

In designing the RVA monitoring within the territory of Slovakia one of the important criteria was a selection of appropriate sites. It was based on an analysis of the results of earlier geological research works dealing with natural radioactivity (e.g. exploration for radioactive raw materials) as well as on newer data and background data obtained from the assessment of geological factors of the environment.

Monitoring of RVA changes in ground air is focused on areas with confirmed elevated (moderate and high) radon risk. Most sites are located in the Spišsko-gemerské rudohorie Mts. area, which has the highest number of sites with high concentrations of soil radon and it is also a region with numerous occurrences of uranium mineralization, including uranium deposits.

The selection of sites for the purposes of soil radon activity measurements on reference areas (RA) was preferably focused on town residential areas: Bratislava (locality Vajnory), Banská Bystrica (Podlavice locality), Spišská Nová Ves (Novoveská Huta and Teplička) and Hnilec.

The soil radon over the tectonics has long been monitored at the Grajnár site, which has been disrupted by the



Fig. 4.2 Situational scheme of monitored objects on the basis of radon risk forecast map (compiled by: A. Gluch & Š. Dzurenda, 2017)
1: Hnilec; 2: Novoveská Huta; 3: Teplička; 4: Podlavice; 5: Vajnory; 6: Dobrá Voda, tectonics; 7: spring Himligárka; 8: spring Zbojníčka; 9: spring Mária; 10: spring of Božena Němcová; 11: spring of St. Andrew; 12: spring area Jašterčie

extensive mining and storage of wood in the locality. Since the 2012 season, the work centre has been moved to the Dobrá Voda area (Trnava District).

The assessment of changes in radon concentrations in groundwater is carried out on objects (available springs, exits, etc.), which have been found in previous works to have increased Rn contents, or high volumetric activities of radon – the area of the Malé Karparty Mts. (springs Mária, Zbojníčka and Himligárka), Bacúch (spring of Božena Němcová), Spišské Podhradie (spring of St. Ondrej [spring of St. Andrew]) and Oravice (the spring OZ – 1).

The monitoring of several sites had been discontinued, interrupted or partially modified as a result of the malfunction of the monitoring sites by unpredictable anthropogenic activities (earthworks, construction, pollution by wild landfills, etc.).

From approximately three tenths of the objects that have been monitored for the past two decades 12 objects have been monitored to-date for soil radon (one of them above tectonic), and six objects for radon in groundwater (Fig. 4.2).

4.3 Methodology

The goal of RVA monitoring in the geological environment is to document and evaluate short-term (seasonal) and long-term (of the order tens of years) changes in radon concentrations in the rock environment (ground air) and groundwater.

A set of geophysical works and activities carried out on objects in the last 15 seasons (period 2002–2016) represents the repeated sampling and measurement of RVA in field and laboratory conditions in the 12 localities (Fig. 4.2) within the territory of Slovakia, including their comprehensive processing, evaluation and alignment of results with previous periods, preparation of individual evaluation reports, updating of resulting databases, etc.

The radon in ground air is monitored at each site within the reference area (RA), which consists of individual points (ground sampling probes) arranged in the profiles, or in an irregular network, up to 400 m². The essential number of points within RA is 17 probes (16 base measurement probes plus one control probe), representing the minimum statistical set to evaluate radon risk RA within each monitoring date.

The ground air for RVA determination is taken through the hand-driven probes from a depth of about 80 cm. The air sample is sucked into the de-emanated and evacuated scintillation Lucas Cell (LC – Calibrated Scintillation Detector, 125 ml volume) which is then transported for measurement in the laboratory conditions. While sampling the ground air besides the data necessary to determine the RVA value, other additional data are being recorded: weather conditions, precipitation, atmospheric pressure, a qualitative assessment of the moisture content of the sediment in the RA area, the drilling/driving resistance of the probes and soil abstraction.

Groundwater samples for the determination of ²²²Rn bulk activity are collected in glass ground-neck samplers, full-filled (volume ca 300 ml), without air bubble. At sam-

pling, the instantaneous value of water and air temperature, source yield, atmospheric pressure, meteorological conditions at sampling, including the data required for RVA calculation, are measured.

Under laboratory conditions, radon from each sampler is bubbled through the washer into four de-emanated and evacuated LCs with a volume of 600 ml which are then measured by a calibrated measuring device in a procedure consistent with RVA measurement in ground air. In order to rule out a random error, two samples are always measured, the resulting radon content being their arithmetic mean. The third sample is analysed, provided the pair of measurements exceeds 10 %.

The method of determination of RVA in ground air and gas permeability of groundwater is determined by approved methodology, which is in compliance with the provisions of Act no. 355/2007 on the Protection, Promotion and Development of Public Health and also in accordance with Decree of MH SR no. 528/2007 laying down the details of requirements for the limitation of exposure to natural radiation.

At RVA determination a calibrated and metrologically validated LUK-4A measuring device (manufactured by RADON v.o.s., Prague, Czech Republic) is used, based on the principle of scintillation detection of alpha particles in Lucas cells.

Measurements of samples of gas mixtures (ground air radon or water sample in admixture with atmospheric air) in the LC are performed under laboratory conditions at 210 minutes after their filling, i.e., after achieving a radioactive equilibrium among ²²²Rn and its decay products.

The volumetric activity of radon (c_A) in ground air is calculated according to:

$$\mathbf{c}_{A} = (Nv - Np)/k \times V \times (3 \times t_{v} \times e^{-\lambda tr})$$
 [kBq x m⁻³]

where: Nv – measured number of pulses of gas mix ture sample in LC for t, time

Np – measured count of LC background pulses for t time

k – coefficient of efficiency of the measuring device

V - sample volume of gas mixture in LC [litres]

t_v - time of gas mixture sample measurement in LC [sec]

t_r - time from ground air intake to LC until start of measurement [min]

λ – decay constant ²²²Rn

The radon risk of RA is assessed in the sense of the amended MoE Directive (currently under approval) and according to Annex no. 7 to Government Regulation no. 350/2006 where the limits for the determination of radon risk categories are recommended based on a quantitative assessment of the measured RVA in ground air and the permeability of soils for gases (Table 4.1).

	3 rd quartile – RA	ADON VOLUMETRIC ACTIVI	TY [kBq x m ⁻³]
RADON		Gas permeability of soil	
RISK CATEGORY	poor	moderate	well
low – I	< 30	< 20	< 10
moderate – II	30 – 100	20 – 70	10 – 30
high – III	> 100	> 70	> 30

Tab. 4.1 Determination of the radon risk category of the reference area

The gas permeability of local soils and rocks is determined for each RA by a granulometric analysis of the sample taken based on the percentage of fine particles **f** (particle diameter <0.06 mm) as presented in the Table 4.2.

Tab. 4.2 Determination of gas permeability of soils and rocks

Permeability	Fine particles proportion	Class in terms of STN 73 1001
poor	f > 65 %	F5, F6, F7, F8
moderate	15 % < f < 65 %	F1, F2, F3, F4, S4, S5, G4, G5
well	f < 15 %	S1, S2, S3, G1, G2, G3

The gas permeability of local soils and rocks is determined for each RA by a granulometric analysis of the sample taken based on the percentage of fine particles **f** (particle diameter <0.06 mm) as presented in the Table 4.2.

Volumetric activity of radon in water (\mathbf{c}_{A}) is calculated according to the relationship:

$$\mathbf{c}_{A} = (Nv - Np)/k \times Vv \times (3 \times t_{v} \times e^{-\lambda tr}) \times e^{-\lambda tF}$$
 [Bq x l-1]

where: Vv – water sample volume in the washer [litres]

 $e^{-\lambda tF} = F(t_F) - a$ coefficient expressing a ^{222}Rn activity decrease over time t_F

Other variables are explained when calculating RVA in ground air.

Decree of the Ministry of Health of the SR no. 528/2007 provides guide values for the implementation of measures (Table 4.3), which are basic indicators when assessing the suitability of the water supplied in terms of natural radioactivity. The parameters set out in this Decree are applied when evaluating the results of RVA determination in waters.

Tab. 4.3 Guide values for the implementation of measures

Type of water supplied	Total volumetric activity alpha [Bq x I ⁻¹]	Total volumetric activity beta [Bq x l ⁻¹]	Volumetric activity ²²² Rn [Bq x I ⁻¹]
Spring water "appropriate for toddlers nutrition"	0.1	0.2	20
Natural mineral water	1.0	2.0	100
Spring water, packed drinking water, potable water	0.2	0.5	100

4.4 Overview and results of realized works

4.4.1 Ground radon on reference areas (RA)

The RVA monitoring in ground air for RA was performed at the given period (2002–2016) at different frequencies at five locations, the position of which is depicted in Fig. 4.2. Overview and comparison of results of the RVA measurements in ground air for individual localities, objects and monitoring seasons is in Table 4.4.

The reference area of Hnilec, located approximately 2.1 km south-east of the centre of the same name on state road no. 533 Spišská Nová Ves – Gemerská Poloma, has long been evaluated in high to extremely high radon risk. The source of soil radon is an enriched medium-coarse-grained *Gemeride* (so-called Hnilec) granite with anomalous uranium content (according to field gamma spectrometry about 20 ppm eU – Čížek, Smolárová & Gluch, 2001). We rank it to the rocks with the highest natural radioactivity in the Western Carpathians.

The monitoring of RVA in ground air here is realized four times a year, every two months, in the early spring to late autumn. The highest mean RVA_{3,Q} (third quartile RVA) values are recorded during the summer/autumn (Fig. 4.3). In October 2005, the highest RVA value in a single probe (1,861 kBq x m⁻³) was measured here, which is the maximum not only for this object, but also – according to the available data – one of the highest values traced on the territory of Slovakia. The RVA in ground air is very varia-

ble $(3-1,861 \text{ kBq x m}^{-3} \text{ in a single probe})$, with a long-term average of 468 kBq x m^{-3} and a standard deviation of 292 kBq x m^{-3} (Table 4.4) of the whole set of measurements (1,018 probes).

The reference area Novoveská Huta is located on the southwest side of the same village along the local communal road (from the church towards a local part Rybníky) in an environment built of colourful sandstones and slates of

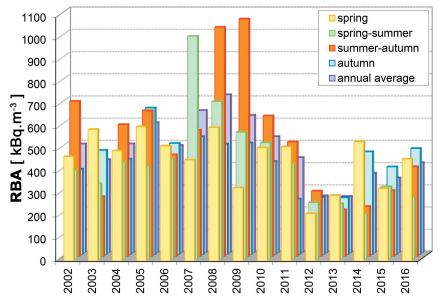


Fig. 4.3 RVA monitoring in ground air, 2002–2016, RA Hnilec (Gluch & Zeman, 2016)

The course of variations of RVA in ground air at the RA Hnilec in individual seasons shows a certain "wavy" character. While RVA correlates well with low precipitation totals (RVA $_{2003}$ = 420 kBq x m⁻³ or RVA $_{2011}$ = 430 kBq x m⁻³), the opposite case is not fully demonstrated. Although the local maximum RVA $_{3,Q}$ from the 2005 season (587 kBq x m⁻³) corresponds to the increased rainfall, but for the exceptionally damp year 2010 this is no longer valid (source: www.shmu.sk; Table 4.4). After the peak in the 2008 season (712 kBq x m⁻³), the RVA $_{3,Q}$ values in the soil in recent years show a rather significant drop to up to 255 kBq x m⁻³ from 2013 (trend RVA $_{2013/2008}$ = 0.36).

the Stráže Mb. of the Novoveská Huta Fm. (Permian) with connection to the NNE – SSW dislocations.

(The monitoring of RVA in ground air is usually carried out 6–8 times during the year, at monthly intervals from the early spring to late autumn periods. During the summer months, high concentrations of soil radon are recorded in this territory (Fig. 4.4). The maximum RVA in a single probe was recorded here in July 2006 (670 kBq x m⁻³).

After the 2006 RVA $_{3,Q}$ peak of 2006 (113 kBq x m⁻³) there was a significant decrease in soil radon concentration to 35 kBq x m⁻³ in the 2013 season (RVA $_{2013/2006} = 0.31$, Table 4.4). However, in a very dry year 2011 (656 mm pre-

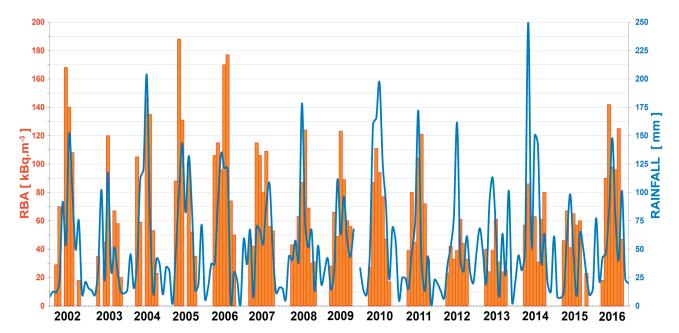


Fig. 4.4 RVA monitoring in ground air, 2002–2016, RA Novoveská Huta (Gluch & Zeman, 2016)

cipitation totals) there was a slight increase in soil radon concentrations (RVA_{3.0} = 71 kBq x m⁻³).

Important and significant results of soil radon monitoring at this locality include the observed phenomenon of a sharp decline in RVA (sometimes up to low risk), which is reflected on RA Novoveská Huta under the same conditions – during the first frosts in the autumn, possibly ground frosts in the spring (with non-frozen soil). Probably as a result of the increased temperature gradient between soil and atmospheric air, the radon leaks more intensely into the atmosphere, resulting in a significant reduction of its content in the cover sediments (Smolárová & Gluch, 2010).

The reference area Teplička is located approximately 2.8 km south of the centre of Spišská Nová Ves in the local part Šulerloch (named after the homonymous altitudinal point 646 m). The RA subsoil is made of Paleogene sedi-

ments (slate, sandstone) with a higher proportion of clay fraction.

Monitoring at this site takes place at monthly intervals between early spring and late autumn. Although the object is monitored on the same day as the Novoveská Huta RA, in a distance of about 5 km, the variations in soil radon concentrations have a significantly different course. Elevated values are mostly recorded in spring and autumn and lows in the summer months (Fig. 4.5). In the single probe, the highest RVA value (196 kBq x m⁻³) was measured in May 2005.

The course of variations of soil radon content in RA Teplička within the individual monitoring seasons has a certain "quasi – sinusoidal" shape, with minimum RVA values correlating with low precipitation totals (RVA₂₀₀₃ = 56 kBq x m^{-3} , RVA₂₀₁₁ = 59 kBq x m^{-3}). After the peak in the 2005 season (92 kBq x m⁻³), the values of RVA_{3,Q} in ground air since 2006 (except exceptionally damp year

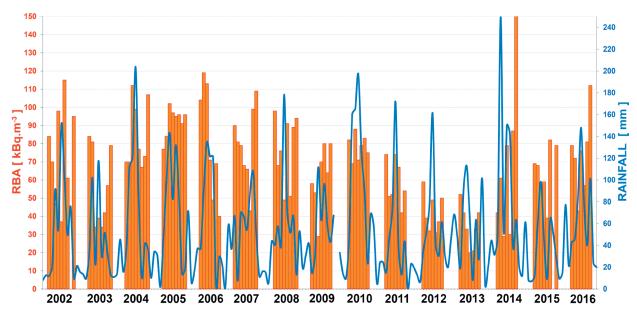


Fig. 4.5 RVA monitoring in ground air, 2002–2016, RA Teplička (Gluch & Zeman, 2016)

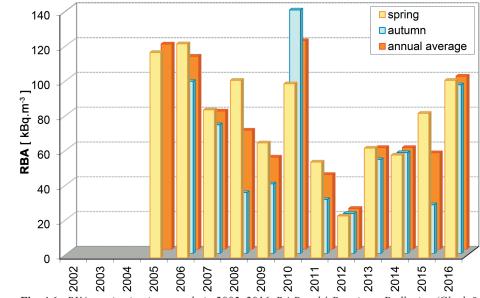


Fig. 4.6 RVA monitoring in ground air, 2005–2016, RA Banská Bystrica – Podlavice (Gluch & Zeman, 2016)

2010) show a rather significant decrease compared to RVA $_{2013} = 35$ kBq x m $^{-3}$ (Table 4.4). Trend RVA $_{2013/2005} = 0.38$.

The reference area Banská Bystrica – Podlavice is based on the NW border of Banská Bystrica (part Podlavice), along both sides of the field road near the gardeners' colony.

The RA lithology forms Ramsau Dolomite with anomalous concentrations of uranium (so-called "uranium dolomites"). The RVA monitoring in ground air was carried out twice during the year (spring and autumn).

Since the 2005 season (RVA $_{3,Q}$ = 118 kBq x m $^{-3}$) – exception is the exceptionally damp season of the year 2010 – this object has manifested a gradual and significant decrease to RVA $_{2012}$ = 24 kBq x m $^{-3}$ (trend RVA $_{2005/2012}$ = 0.20), observable especially in autumn monitoring (Fig. 4.6). The highest radon content in the ground air in a single probe (272 kBq x m $^{-3}$) was found in May 2006.

The reference area Bratislava – Vajnory is situated on the north-eastern edge of a homonymous part of Bratislava, along the meliorating channel of approximately N – S direction.

The lithology of the object form relatively well gas-permeable fluvial sediments. Monitoring of RVA in ground air in the area of the RA Bratislava – Vajnory is realized from the 2005 season twice a year (spring and autumn). In the period 2005–2011 (with the exception of the exceptionally wet season 2010), the average annual levels of RVA (Fig. 4.7) decreased gradually and significantly (RVA_{2011/2005} = 0.46). In a single probe, the maximum content of soil radon (122 kBq x m⁻³) was recorded in May 2005.

Limit levels of RVA $_{3,Q}$ in ground air were recorded here in the seasons 2010 (69 kBq x m⁻³), or 2012 (19 kBq x m⁻³), trend RVA $_{2012/2010}$ = 0.28.

In the 2013 season, RVA measurements in the ground air were performed on the DV–2 geophysical profile, located about 80 m NW from the DV–1 profile. The gasometry was performed simultaneously with electrical resistivity tomography (ERT) with a 5.5 meter measurement step. The monitored failure zone was also indicated on this profile by a significant increase in soil radon concentrations with RVA_{MAX} = 20 kBq x m⁻³ compared to the normal range of approximately 1 kBq x m⁻³, as well as by a significant drop in the resistance level from over 3,000 ohm.m in poorly disintegrated Wetterstein Dolomite up to a value below 300 ohm.m above the tectonic dislocation.

In the 2014 and 2015 seasons, detailed measurements of soil radon concentrations on P– or P–2 (each with 6 emanometric profiles, network of 5 x 10 m), located in the vicinity, or south of the emanation anomaly traced in 2012 at the DV–1 profile. The disturbance zone on the P–2 area was manifested by anomalous increase of the soil radon concentrations RVA $_{\rm MAX}=48~{\rm kBq}~{\rm x}~{\rm m}^{-3}$, in the normal range of approximately 7 kBq x m $^{-3}$.

In season 2016, a detailed radon survey continued SSE towards P–3 area – five emanometric profiles in the 2–5 x 5–10 m (according to terrain accessibility). The monitored disturbance zone has been indicated to date by the highest soil radon content – RVA $_{MAX}$ = 62 kBq x m⁻³, in the normal field below 10 kBq x m⁻³.

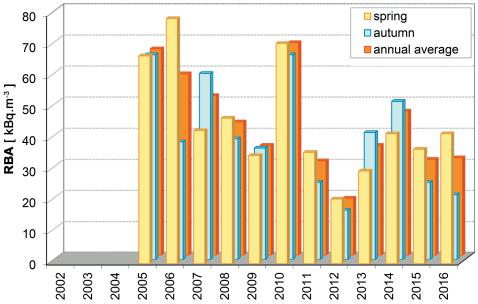


Fig. 4.7 RVA monitoring in ground air, 2005–2016, RA Bratislava – Vajnory (Gluch & Zeman, 2016)

4.4.2 Ground radon on tectonics

The first indicative emanation measurements at the **Dobrá Voda site** were realized in the 2012 season on the DV-1 profile, based approximately 1.1 km NNW of the centre of the Dobrá Voda Village. The profile was of approximately NE – SW direction, of a length of 80 m, picking step 5 m (Fig. 4.8).

The failure zone, observed in Wetterstein Dolomite (Middle Triassic), covered by fluvial flood-plain sediments, resulted in a significant increase in soil radon concentration with RVA $_{MAX} = 24 \text{ kBq x m}^{-3}$ compared to the normal field below 2 kBq x m^{-3} .

The area of the failure zone in the Dobrá Voda area (Fig. 4.8) is clearly manifested by the increase of the concentrations of radon in the ground air. The resulting tectonic dislocation positively affects the transport of radon to the subsurface parts even from greater depths, so that the soil radon contents reach anomalous values, radically exceeding the background.

After obtaining the necessary instrumentation for the continuous measurement of the RVA and the construction of a monitoring facility (observation well), we assume the future interconnection of PMSGF subsystems 05 (radon volumetric activity) and 02 (Tectonic and seismic activity of the area).

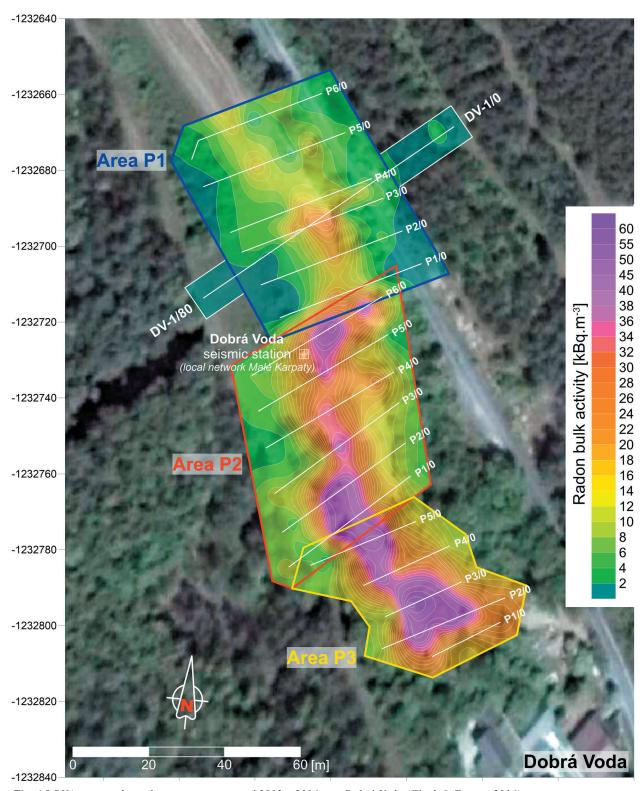


Fig. 4.8 RVA in ground air above tectonics, period 2012 – 2016, site Dobrá Voda (Gluch & Zeman, 2016)

4.4.3 Radon in groundwaters

Radiohydrochemical sampling and monitoring of RVA in groundwater was carried out at different frequencies in six locations (Fig. 4.2) between 2002 and 2016. The overview and comparison of radon concentration measurements in groundwater by individual localities, objects and monitoring seasons is documented in Table 4.4.

In the area of the Malé Karpaty Mts. (about 9 km north of the centre of Bratislava), the sources of Himligárka,

Zbojníčka and Mária (spring of Mary) are monitored twice a year (spring and autumn). Captured and reconstructed springs are related to the acidic environment of the Malé Karpaty Mts (crystalline (leucocrate, muscovite and two-mica granites, granodiorites, of the Bratislava type).

The groundwaters here have a shallow circulation with a link to the zone of disintegration in which conditions for the formation and spread of radon are favourable.

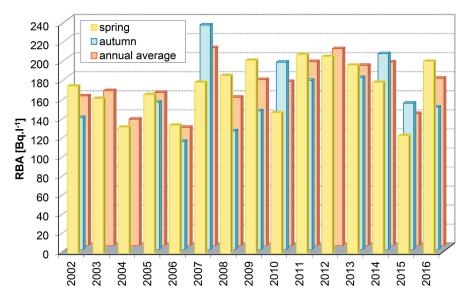


Fig. 4.9 RVA monitoring in groundwater, period 2002–2016, spring Himligárka (Malé Karpaty Mts.; Gluch & Zeman, 2016)

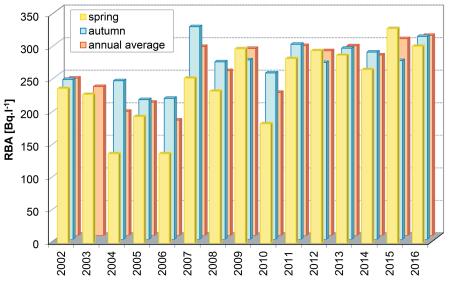


Fig. 4.10 RVA monitoring in groundwater, period 2002–016, spring Zbojníčka (Malé Karpaty Mts.; Gluch & Zeman, 2016)

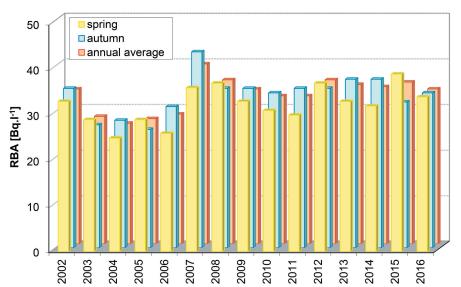


Fig. 4.11 RVA monitoring in groundwater, period 2002–2016, spring Mária (Malé Karpaty Mts.; Gluch & Zeman, 2016)

The spring of Himligárka is situated about 30 m from the so-called. Štefánikova magistrála (important tourist route through Slovakia, labelled in red) and about 600 m NNE from the Horný Červený Kríž altitudinal point.

Increased concentrations of ²²²Rn are recorded mostly in the spring; decreased in autumn. However, the highest radon content (236 Bq x l⁻¹) was measured in the 2007 autumn measurement (Fig. 4.9). The yield of the spring varies widely between 0.01 and 1.86 l x s⁻¹ while in the autumn of 2004 and 2012 the spring was even dried.

The spring Zbojníčka is situated about 800 m from the source of Himligárka and about 270 m west of the Horný Červený Kríž altitudinal point.

As a rule, higher volumes of radon activity are recorded in autumn, with a maximum RVA (330 Bq x l⁻¹) found in the spring of 2015 (Fig. 4.10). The yield of the spring is varied in the range of 0.04 to 3.33 l x s⁻¹, while the spring was dried in 2003.

At the spring of Mária, about 2 km southwest of the Zbojníčka spring, there are long recorded relatively low contents of ²²²Rn, in the long-term average slightly higher in autumn monitoring. The highest RVA level (43 Bq x l⁻¹) was measured in the autumn 2007 (Figure 4. 11). The yields of the source fluctuate in the range of 0.03 to 1.82 l x s⁻¹.

The spring of Božena Němcová, located approximately 1.4 km north of the centre of Bacúch (Brezno District), was monitored 6–8 times a year in the period under review. Captured and engineered spring object emerge to the surface in the environment of garnet two-mica gneisses and mica schists. The radon is bound here to fault tectonics and to disintegrated, emanation-positive zones of the geological environment.

In the water of the given source were found increased to high levels of radium ²²⁶Ra (monitored in the period 1998–2000), which were not stable and varied in the range of about 0.5–1.8 Bq x l⁻¹ without correlation to ²²²Rn concentrations in water.

The radon content during the last 15 seasons was very variable and ranged from 165 to 422 Bq x l⁻¹, the highest value being measured in April 2008 (Fig. 4.12). The varia-

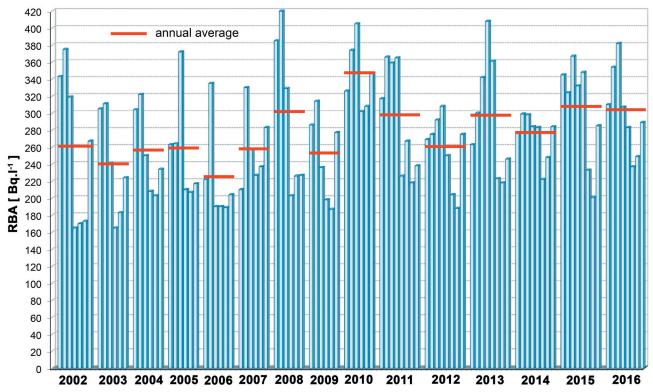


Fig. 4.12 RVA monitoring in groundwater, period 2002–2016, spring of Božena Němcová (Bacúch; Gluch & Zeman, 2016)

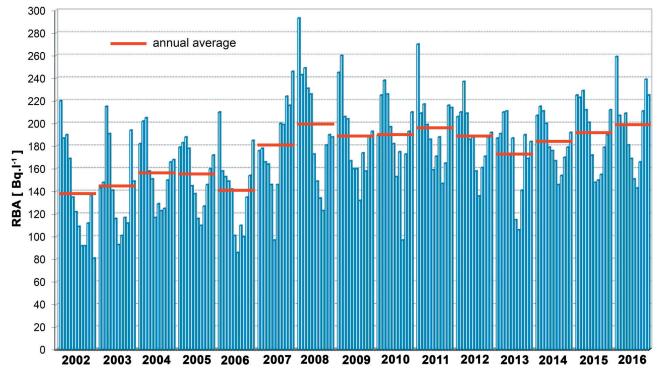


Fig. 4.13 RVA monitoring in groundwater, period 2002–2016, spring of St. Andrew (Spišské Podhradie; Gluch & Zeman, 2016)

tion graph has a relatively regular "sinusoidal" course with RVA maxims at the end of winter (February – April) and minima in summer and autumn months (July – October). The yields of the spring are low but relatively stable in the long run and range from 0.02 to 0.03 l x s $^{-1}$.

The mineral spring of St. Andrew at Spišské Podhradie, is situated in the area of Sivá Brada on the State Road no. 18 (E 50) Poprad – Prešov. Captured and engineered spring object emerging from travertine mound in an area built of clayey-stony colluvial sediments is monitored 12 times a year at monthly intervals.

Its waters have a deeper circulation and therefore, besides radon, they also have an increased content of ²²⁶Ra.

We assume that the source area for the radioactivity of these waters is in the base of Triassic carbonates. Like $^{222}\rm{Rn}$ in water, also $^{226}\rm{Ra}$ concentrations were not stable in the water and during the 1998 – 2001 season they varied between 0.2–1.8 Bq x l¹¹ without correlation to radon contents.

The volumetric activities of radon in the period under review ranged from 81 to 293 Bq x l⁻¹ (Table 4.4). The highest RVA was recorded in January 2008 (Fig. 4.13). The RVA variation curve has a relatively regular "wave" pattern that repeats throughout the monitored period. Every year, in the winter and early spring (January – April), measured volumes of ²²²Rn volumetric activity reached up to about three times its minimum contents, measured in

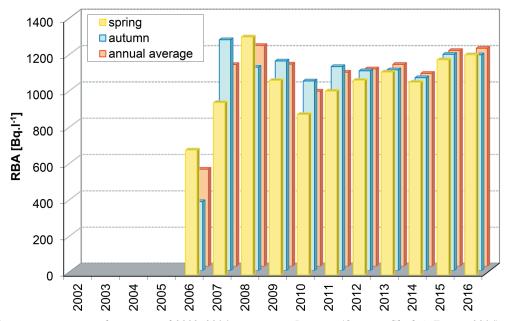


Fig. 4.14 RVA monitoring in groundwater, period 2002–2016, spring area Jašterčie (Oravice; Gluch & Zeman, 2016)

summer to autumn (June – September). The yields of the spring are relatively low but stable and in the period under consideration they varied in the range of $0.01-0.06 \, l \, x \, s^{-1}$.

The Jašterčie spring area, located about 1.8 km south of Oravice, is monitored twice a year (spring and autumn). The spring is located about 30 m from the OZ-1 borehole, which (probably due to corrosion and pressure) in 1999 unplugged the shutter at the initial casing section of the borehole, followed by a strong water outlet directly into the spring area. Thanks to this accident, he monitoring had been interrupted until 2006.

The waters of the source in question have a deep circulation, bound to the Pre-Tertiary formations and deep tectonic dislocation zones. Source of ²²²Rn in groundwater is not only emanative (originating from the environment of emitting rocks), but also autogenic (the resulting dispersion of radium contained in water). The radium contents were monitored by 2000 (volumetric activity of ²²⁶Ra was 0.5–1.7 Bq x l⁻¹).

In this natural thermal exsurgence of groundwater – according to available information – the highest RVA in natural groundwater is reached within the entire territory of

Slovakia. This is confirmed by the results of the measurements in which the RVA ranged from 382-1,312 Bq x 1^{-1} (Fig. 4.14). Up to now the highest concentration of 222 Rn in water -1,407 Bq x 1^{-1} here was recorded in autumn monitoring in 1998.

Due to the nature of the object – the source with the aged damaged collector object – it is not possible (without relatively difficult technical works) to determine its yield.

4.5 Conclusions

The results of the monitoring of RVA variations in ground air have long been confirmed by the fact that radon concentrations are significantly dependent on meteorological and climatic factors and thus on the moisture and gas permeability of local soils and rocks but to a negligible extent also on the structural and geological setting itself and the lithological characteristics of the rock environment areas of a specific location.

The meteorological conditions in the last 15 monitoring seasons were considerably different for the accumulation and possibilities of radon spreading in the rock environment. It is generally assumed that the moisture content of the cover deposits "positively" influences radon concen-

Tab. 4.4 Course of values of radon volumetric activity (RVA) in ground air and groundwater, period 2002 – 2016 (Gluch & Zeman, 2016)

3	GILL				1				Year				[Long-	3	
		2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	average	•	Z
	Ground radon at reference areas								3.0	c _A [kBq x m ⁻³]	x m ⁻³]								
_	Hnilec	491	420	491	587	485	642	712	620	524	430	258	255	359	334	407	468	292	1,018
7	Novoveská Huta	68	58	87	66	113	73	61	29	99	71	39	35	09	51	88	70	29	1,767
3	Teplička	92	99	80	92	81	62	77	62	78	59	42	35	89	64	74	89	31	2,372
4	Bratislava – Vajnory				29	69	52	43	36	69	31	19	36	47	31	32	43	20	408
S	Banská Bystrica – Podlavice				118	111	08	69	53	120	43	24	59	59	99	100	74	51	391
	Ground radon on tectonics																		
_	Dobrá Voda, profile DV-1											9		*	(*		9	292	18
2	Dobrá Voda, profile DV-2												3		(*		3	29	09
3	Dobrá Voda, area P-1													10	(*)	10	31	09
4	Dobrá Voda, area P-2														19	(*	19	20	09
5	Dobrá Voda, area P-3															36	36	51	37
	Mean precipitation totals in Slovakia	861	573	851	938	922	894	098	068	1,255	959	747	864	957	719	924	[mm]		
	in [mm] and [%] of long-term mean	106	75	112	125	101	122	112	122	157	80	86	122	119	86	124	[%]		
									Year								Long-		
Nr.	SILE	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	term average	ò	Z
	Radon in water								c	_A [Bq x l ⁻¹]	[1-1]								
1	Bratislava – spring Mária	34	28	27	28	29	40	36	34	33	33	36	35	35	36	34	33	4	09
2	Bratislava – spring Zbojníčka	242	230	161	205	178	167	254	288	220	294	284	292	278	303	308	257	20	09
3	Bratislava – spring Himligárka	158	163	133	161	125	208	156	175	173	194	207	190	193	139	176	170	32	54
4	Bacúch – spring of Božena Němcová	259	238	254	256	222	257	299	250	344	295	258	295	274	305	301	274	63	206
5	Spišské Podhradie – spring of St. Andrew	137	143	156	154	140	180	198	187	188	195	187	172	183	191	197	174	41	365
9	Oravice – spring area Jašterčie					536	1,112	1,217	1,115	996	1,070	1,088	1,113	1,064	1,190	1,202	1,061	205	45

mean value of the third quartile RVA in ground air for assessed year 3.Q cA Note:

mean value of RVA in groundwater for assessed year cA

monitoring was interrupted for the given period of time

*

standard deviation of RVA from all measurements at the site through the entire monitoring period

number of RVA measurements at the site through the entire monitoring period the object was not monitored in the given year οZ

trations in the ground air, because the higher humidity of the rock environment more or less effectively slows the penetration of radon to the surface and further into the air. This leads to an increase of its contents in the ground air and vice versa — when the soil humidity decreases, RVA values also decrease. This dependence has manifested itself to a large extent on most monitored sites. The relationship between raising the RVA level in ground air in high precipitation means is unclear, and it will require a longer-term monitoring,

Seasonal variations of RVA in ground air depend not only on the moisture and gas permeability of local soils and rocks, but also on the geological setting itself and the lithological characteristics of the particular site. It follows that, even under the same meteorological conditions, but in different geological settings, the character of variations may not be the same. This knowledge is one of the significant findings in monitoring the variations of RVA in ground air within the geological task solution.

An example of this are the results of the RVA monitoring in the ground air at RA Novoveská Huta and RA Teplička (Paleogene sediments with medium to low gas permeability, with an increased proportion of clay fraction) which are relatively close to each other (about 5 km) practically in the same climatic area but with a different geological profile in which the accumulation and spread of radon are monitored.

Both of these sites were mostly monitored on the same day (i.e. in comparable meteorological conditions) but the results of RVA measurements in ground air show a different course – in the summer months the RA Novoveská Huta recorded increased and the RA Teplička reduced radon content in soil and the opposite behaviour in spring and autumn (Novoveská Huta low and Teplička high RVA).

Among the significant results of the monitoring of radon content in ground air should be included the observed phenomenon of a sharp decline in RVA, which is reflected in RA Novoveská Huta during the first frosts in the autumn, or even ground frosts in the spring. Obviously, due to the temperature gradient between soil and atmospheric air, radon more intensely leaks into the atmosphere and its content in the cover deposits decreases.

The results of RVA monitoring in ground air have long documented the variability of its contents in the surface parts of the rock environment during the year but also over the monitored seasons. The relatively significant dependence of RVA levels on meteorological conditions but with an ambiguous effect on individual localities is confirmed, which is evidently due to their different structural geological and lithological characteristics.

Variations in radon volumetric activity in monitored groundwater sources are of rather seasonal nature and, during monitoring over several seasons, show a certain wavy, "sinusoidal" regularity. Unlike soil radon, they are not so much influenced by random phenomena, changes in the atmosphere and are not so "sensitive" to various short-term weather changes (temperature, atmospheric pressure). Maximum levels of RVA in groundwater are generally recorded in winter and/or in spring and minimum

values in summer and autumn months. Neither correlation between ²²²Rn and ²²⁶Ra volumetric activities has been demonstrated on any of the monitored objects, nor was the correlation between source yield and concentration of radon in groundwater samples taken.

Observed tectonic failure zones (localities – Dobrá Voda, Grajnár) are clearly manifested by increasing concentrations of soil radon. The mapped dislocations positively affect the transport of radon to the surface parts even from greater depths, so that the soil radon contents reach anomalous values. From a practical point of view this can be of interest, for example, when observing "active" tectonic dislocations, when there is a high probability of their positive identification even under less favourable conditions.

From the practical point of view, the knowledge gained is of far-reaching importance, because they may lead to considerable underestimation of the radon risk at the building site due to measurements made under inappropriate meteorological conditions (long-term drought, periods with high precipitation, significant temperature differences between the atmosphere and the sediments mainly in the spring, late autumn or winter). A thorough assessment of the geological environment (especially in stratified sediments with increased proportion of clay fraction, the presence of tectonic dislocations, etc.) is also necessary in evaluating the results of these measurements.

The evaluation of the RVA monitoring results in the geological environment is documented by the fact that the variations in its concentrations are both regular (seasonal) and also random (local, time, ...). Gradually collected knowledge on the variability of radon content in rock and groundwater, their evaluation, processing, and availability of monitoring results through Internet services are clearly beneficial to a more objective assessment of radon risk from a geological environment.

More reliable results can be obtained by processing and evaluating long-term monitoring systems, outputs of which can provide relevant background for adopting more general conclusions in this area. This intention is also followed by the implementation of the PMS GF project at the SGIDŠ.

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