

Uranium occurrences as landfill residuum after closed mining in the Kravany area (Kozie chrby Mountains, Western Carpathians, Slovakia)

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Abstract: The Kozie chrby Mountains are known for their potential for uranium deposits and mineral occurrences, many of which have been mined and exploited in the past. Pilot ground gamma-ray spectrometry measurements along six profiles crossing the Vikartovce fault map trace were carried out in the north-east vicinity of the village of Kravany to specify its position in the field. Extremely high radioactivity values were measured especially the absorbed dose rates and uranium concentrations. The results of an additional eleven rosette-shaped profiles at the three most radioactive sites confirmed the high values. The average values of the absorbed dose rate (136.4 nGy.h^{-1}) and the uranium concentration (8.8 ppm eU) in the studied location greatly exceeded the average values for the rocks of the Western Carpathians, 63.3 nGy.h^{-1} and 3.3 ppm eU , respectively. The maximum measured values of $1482.6 \text{ nGy.h}^{-1}$ and 223.9 ppm eU were on the order of 10- to 100-times higher than the average values. In the case of potassium and thorium, the average values were exceeded only rarely and not as significantly. The cause of the increased and high values of radioactivity of the rock and soil environment are the remnants of uranium mining as well as the reclamation of mining facilities and activities. The radioactively contaminated near-surface horizon over an area of more than 2000 square meters of the studied location may pose a significant health risk to people potentially staying in the area during the annual cycle. This information should be evaluated when planning future human activities in the area studied.

Key words: Western Western Carpathians, Kozie chrby Mts., Permian, uranium deposit, landfill, remediation, gamma-ray spectrometry

1. INTRODUCTION

Thanks to the rich diversity of the geological structure of the Western Carpathians, Slovakia is among those countries where relatively rich deposits and occurrences of uranium are found. Looking for and the exploring of uranium ores have been carried out in all geological units of the Western Carpathians. Such work has concentrated mainly on the crystalline massifs of the core mountains, the Permian formations throughout Slovakia, the Neogene volcanic mountains and the Tertiary basins. Geological exploration and uranium mining in Slovakia after World War II were induced by strategic objectives. The prospecting and research of uranium mineralization started in the 1950s. The first known uranium deposit was found in Novoveská Huta near Spišská Nová Ves. The study of uranium mineralization continued in the early 1970s in the Permian rocks of the Kozie chrby Mts., in the uranium deposits of Kravany, Vikartovce, Švábovce and Spišský Štiavnik. Later, the survey continued in the Permian rocks of the Považský Inovec Mts., in the uranium deposits of Kálnica and Selec. More extensive uranium mineralization was also found underlying the known mineralized horizon in Novoveská Huta and other localities during the 1970s. Study was renewed during the 1980s, and new deposits were found in the Permian rocks

of the eastern Slovakia. The last important U-Mo deposit was found at Jahodná, near Košice. The uranium mineralization in the Western Carpathians is bound mostly to the Permian rocks and less to the Early Paleozoic rocks. Insignificant occurrences are known also in other formations (Rojkovič, 1997). There are eleven uranium deposits in Slovakia (Figure 1A), two of which are major ones (Novoveská Huta, Jahodná); the remaining nine are small ones. Three of the small deposits have been mined out completely (Muráň, Kravany, Vikartovce), and five others partly (Novoveská Huta, Švábovce, Nižný Chmelienec, Kálnica, Selec). Two deposits have never been mined (Jahodná, Spišský Štiavnik). All the above-mentioned deposits consist of bedded orebodies of lenticular or irregular shape. Disseminated ore minerals are bound to carbonaceous flora remains in sandstones (Kravany, Švábovce). Elsewhere, the ore minerals are disseminated in volcanoclastic sediments (Muráň, Novoveská Huta, Kálnica), in volcanic bodies (Jahodná), and on faults (Novoveská Huta, Kravany) (Daniel et al., 1996). While between 1947 and 1989 radiometric investigations were aimed at the prospecting, exploring, and mining of uranium deposits, since 1990 radiometry has mostly been used to resolve environmental issues – to measure radioactivity from an environmental point of view and to compile maps of environmental factors. Such statewide projects were carried out as the Research of

Geologic Environmental Factors (1991 – 1995) (Daniel, 1999), the Derived Map of Radon Hazard (1991 – 1995) (Čížek et al., 1995), the Atlas of Geophysical Maps and Profiles (1998 – 2001) (Čížek et al., 2001), the Maps of Radioactivity of ^{137}Cs (1997 – 2005) (Gluch et al., 2005) and the project for finishing works on radioactive ores mining “Revision and finishing works on radioactive mineral materials” (1995 – 1999) (Daniel, 1999). Neither uranium mining nor exploration are currently taking place in Slovakia (Daniel et al., 1996).

EXPERIMENTAL PART

The article focuses on the geophysical exploration of the radioactivity of the ground rock and soil environment as well as the analysis, presentation, and interpretation of the measured results. The main aim is to point to the existence of easily accessible surface places with high natural radioactivity, in this case as the result of former uranium mining activity, and to compare the radioactivity composition and its level with generally acceptable limits. The area of study is localized in the close vicinity of Kravany, a village on the southern slopes of the Kozie chrbty Mts., where a former uranium mine once operated (locality No. 5 in Figure 1A). High values of natural radioactivity were found here during the Vikartovce fault study within the ambit of the Slovak Research and Development

Agency project “Multidisciplinary research of geophysical and structural parameters, and environmental impacts of faults of the Western Carpathians”, and they were attributed to the fault.

GEOLOGICAL SETTING

The main geological units of the Kozie chrbty Mts. region are the Alpine-folded Permian, Triassic and unconformable overlying Paleogene (Figure 1B). The Permian is represented by the Malužiná Formation (Vozárová & Vozár, 1988), up to 2.2 km thick, which is a clastic sedimentary formation with sporadic chemical sediments. Sandstone is the dominant sediment there, and andesite-basalt volcanites and accompanying volcanoclastics are frequent. Lithology is characterized by three megacycles with sediments that are fine grained upwards. The lower part of each megacycle consists of conglomerates and coarse-grained sandstones. Sandstone and shale dominate in the middle part. Chemical sediments, such as dolomite, dolomite limestone and scarce gypsum with shale and sandstone, form the upper part of the megacycles (Rojkovič, 1997). Mineralization is situated in the pelitic-psammitic Kravany Beds 175–300 m thick (Novotný & Badár, 1971). These beds are present in the middle and upper part of the second megacycle (Vozárová & Vozár, 1988). Ore-bearing rocks are represented by fine-grained

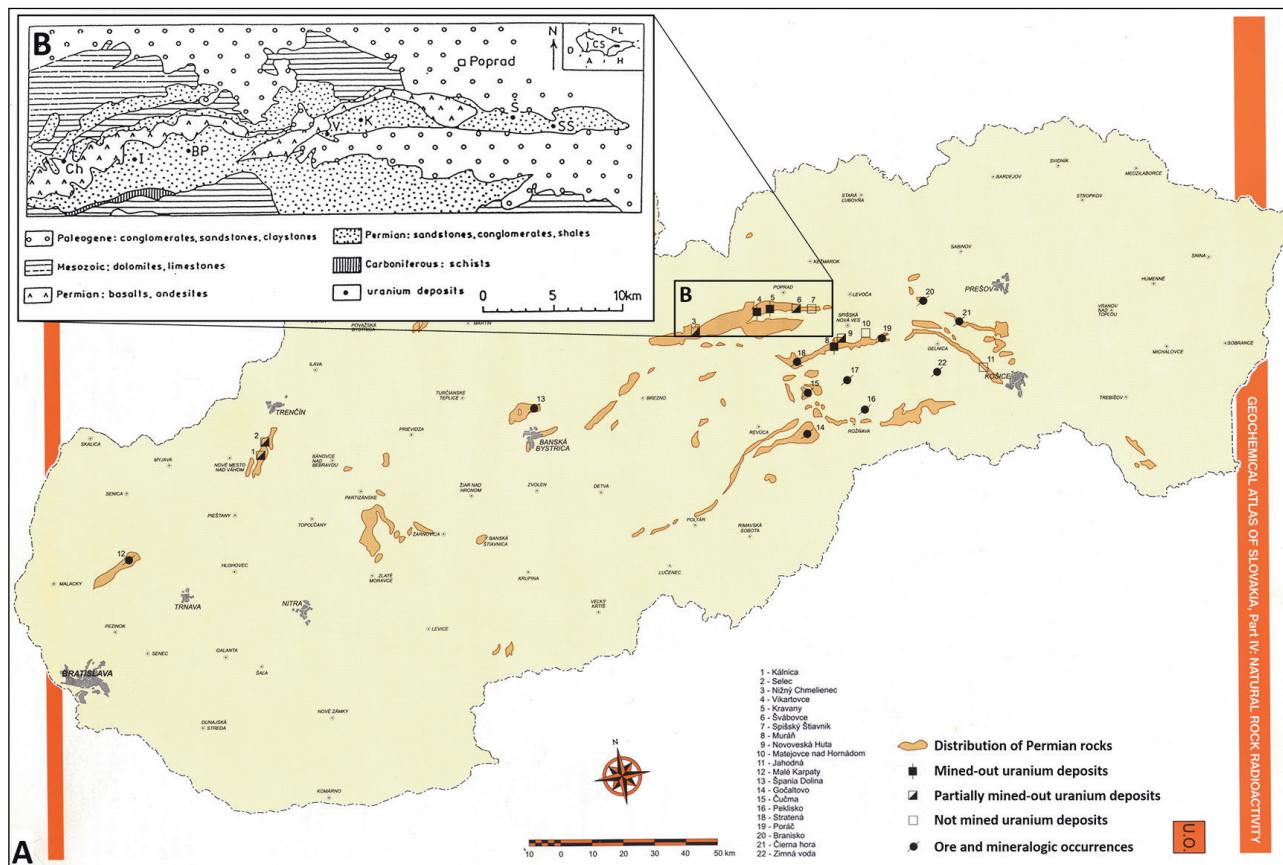


Figure 1 A – Map of uranium ores distribution in Slovakia (Daniel et al., 1996), B – Location of uranium deposits and occurrences in the Kozie chrbty Mts. (adapted after Bystrický et al., 1963 in Rojkovič, 1997): K – Kravany, SŠ – Spišský Štiavnik, Š – Švábovce, V – Vikartovce, and Nízke Tatry Mts. area: BP – Benkovský potok, CH – Nízny Chmelenc, I – Ipolitica.

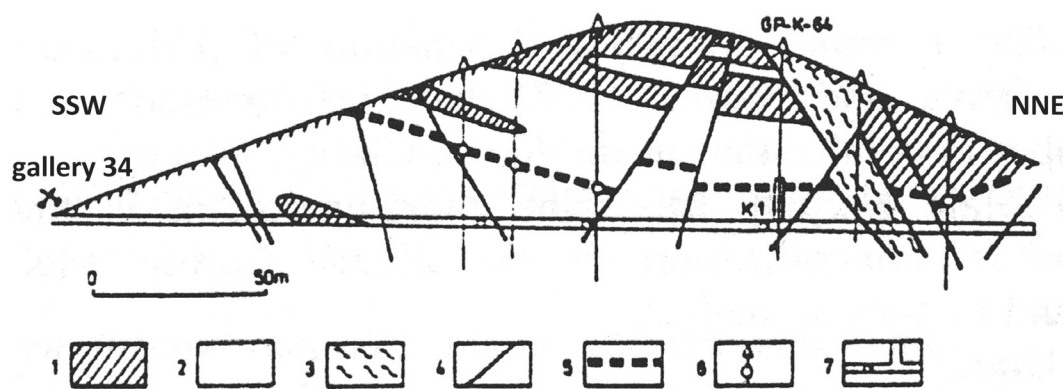


Figure 2 Geological section of the Kravany deposit (according to Veselý & Badár, 1984 in Rojkovič, 1997): 1 – gray green and brown red sandstone, 2 – gray to gray black arkosic sandstone, 3 – tectonic zone, 4 – fault, 5 – ore horizon, 6 – drilling, 7 – mining.

arkosic sandstones and arkoses. Granitoids, migmatites, and high-grade metamorphosed crystalline gneisses were the main source of clastic material (Vozárová & Vozár, 1988).

The Permian sequence is overlain by the Lower and Middle Triassic sediments, represented by sandstone, shale, and mainly by the Middle Triassic dolomite and limestone.

The Paleogene basal transgressive beds are dominated by breccia, conglomerate, and greywacke (Marschalko et al., 1966 in Rojkovič, 1997). They also contain lignite in the studied area.

The structure of the region is affected by lithofacies development, the Alpine nappe, and younger fault tectonics. Significant morphological features were attained by uplifts and

erosion during the Pliocene and the Pleistocene (Lukniš, 1959 in Rojkovič, 1997).

Uranium mineralization in the Kozie chrby Mts. can be observed in the Vikartovce, Kravany, Švábovce, and Spišský Štiavnik deposits (Figure 1B). An ore-bearing horizon from 25 to 75 m thick occurs in the upper part of the Kravany Beds (Figure 2). Gray arkosic sandstone with abundant plant debris is mineralized. The ore bodies form irregular lenses of small thickness, mostly of 1 m (Veselý & Badár, 1984 in Rojkovič, 1997). Uraninite is the main ore mineral and is frequently accompanied by pyrite. The highest concentration of ore minerals was found in the central parts of the lenses, particularly in 1–3 cm thick coal layers (Rojkovič, 1997).

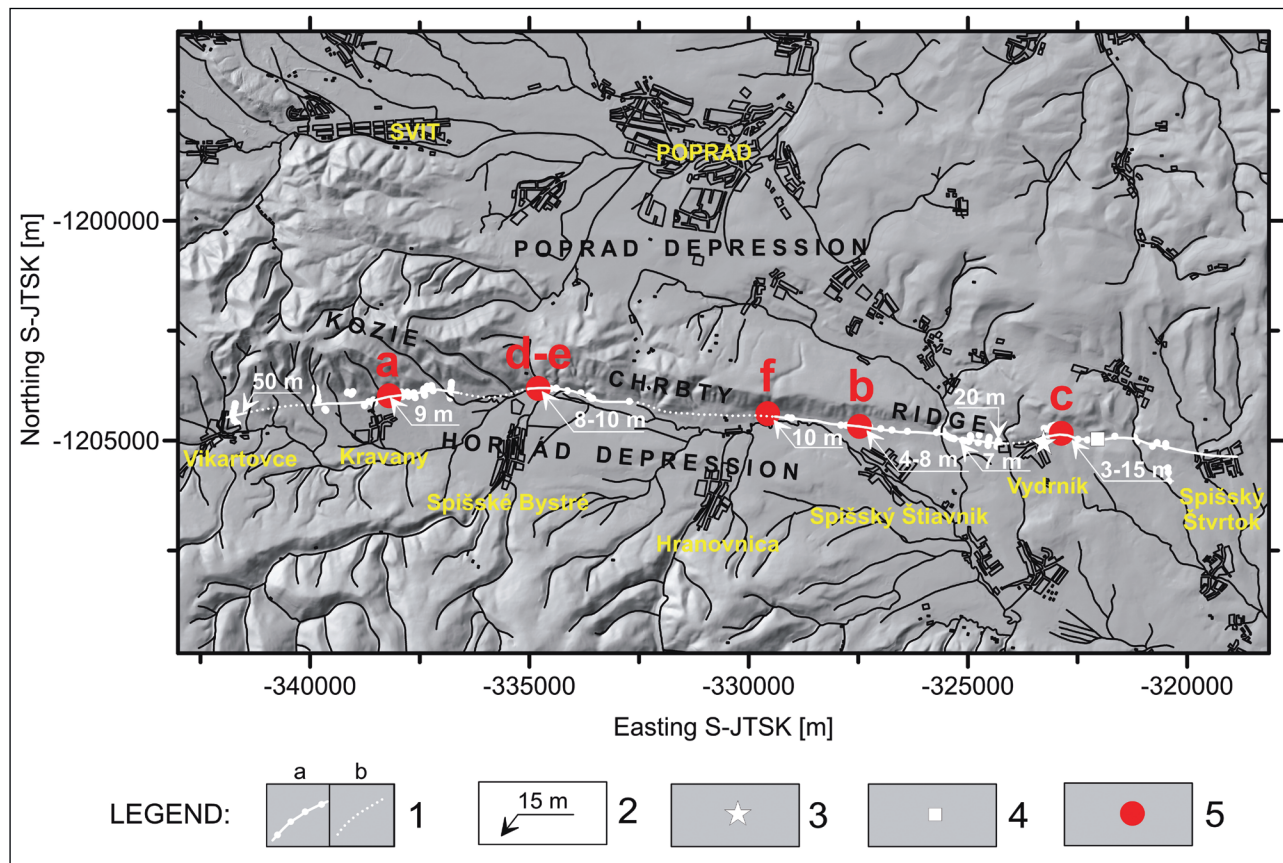


Figure 3 The Vikartovce fault (VIF) trace and localization of geophysical measurements (Marko et al., 2022) on a topographic map (GCI & NFC, 2017-2019): 1 – VIF trace detected and mapped (a), estimated (b); 2 – width of the damaged zone; 3 – the highest indoor radon concentration in the village Vydrič; 4 – occurrence of travertine; 5 – survey profiles localization: a – Kravany, b – Spišský Štiavnik, c – Vydrič, d and e – Spišské Bystré, f – Hranovnica-Dubina.

METHODS USED

A proven package of ground geophysical survey methods with shallower or deeper measurement range was used to study the location, route, and some physical properties of the Vikartovce fault as the dividing line between the Kozie chrbty Mts. horst and the Hornád Depression (Marko et al., 2022) (Figure 3). The information on ground radioactivity was taken using the gamma-ray spectrometry method, which provides valuable information on 4 quantities of gamma radiation from rock and soil. This is achieved with a portable 256-channel spectrometer with a 0.347-liter volume NaI(Tl) scintillation detector and a 12-channel embedded GPS receiver (Pico Envirotec, 2014). These quantities were as follows: the total gamma-ray activity in the form of the gamma absorbed dose rate in the air at 1 m elevation above the ground \dot{D}_a (in nanograys per hour [$\text{nGy}\cdot\text{h}^{-1}$]) and the ^{40}K , ^{238}U , and ^{232}Th concentrations in the near-surface horizon. These latter measurements were recorded in percentage K, ppm eU and ppm eTh. A Bluetooth-connected handheld system controller and data logger enabled a walking-mode survey with a 1 Hz measuring frequency and approximately 2 m sampling distance. The profile measurement provides curves along the studied profile crossing the fault line, and the method is sensitive to the local litho-geochemical composition, which specifies the geological zone. Measurements were performed using a Pico Envirotec PGIS-2 system in walking mode.

RESULTS AND DISCUSSION

Ground gamma-ray spectrometry was usually among the first geophysical methods used to identify the route of a geological fault or fault zone during our fault research in the Western Carpathians (Marko et al., 2022). This method is not able directly detect the fault location, but through its densely measured ground radioactivity data it can offer very valuable information on the lithological structure of the near-surface soil and rock layer and thus determine some faults that function as a lithological boundary. Field walking gamma-ray spectrometry was performed in the vicinity of Kravany (locality (a) in Figure 3) on the border between the southern slopes of the Kozie chrbty Mts. and the Hornád Depression, trying to cross the Vikartovce fault line as drawn in Google Earth map (Google Earth Pro, 2025) and

on a geological map (Geological map of Slovakia M 1:50 000, online, 2013) (Figure 4).

In total, six survey profiles were measured with a sampling density better than 1.4 m in walking mode. Based on the categorization of natural radioactivity levels enacted by Regulation of the Ministry of Environment of the Slovak Republic No. 1/2000-3 for compiling and editing maps of natural and artificial radioactivity on a scale of 1:50 000 (low level up to $40 \text{ nGy}\cdot\text{h}^{-1}$, medium level $40 - 80 \text{ nGy}\cdot\text{h}^{-1}$, increased level $80 - 220 \text{ nGy}\cdot\text{h}^{-1}$, and high level over $220 \text{ nGy}\cdot\text{h}^{-1}$), the results of the absorbed dose rate \dot{D}_a , along the measured lines are presented in Figure 4. Four sites with the highest category of natural radioactivity of rocks and soils of over $220 \text{ nGy}\cdot\text{h}^{-1}$ were found on profiles PF3, PF4, PF6, and PF7 (red color). Their radioactive characteristics are presented in Table 1.

Table 1 Sites with the highest natural radioactivity of rocks and soils.

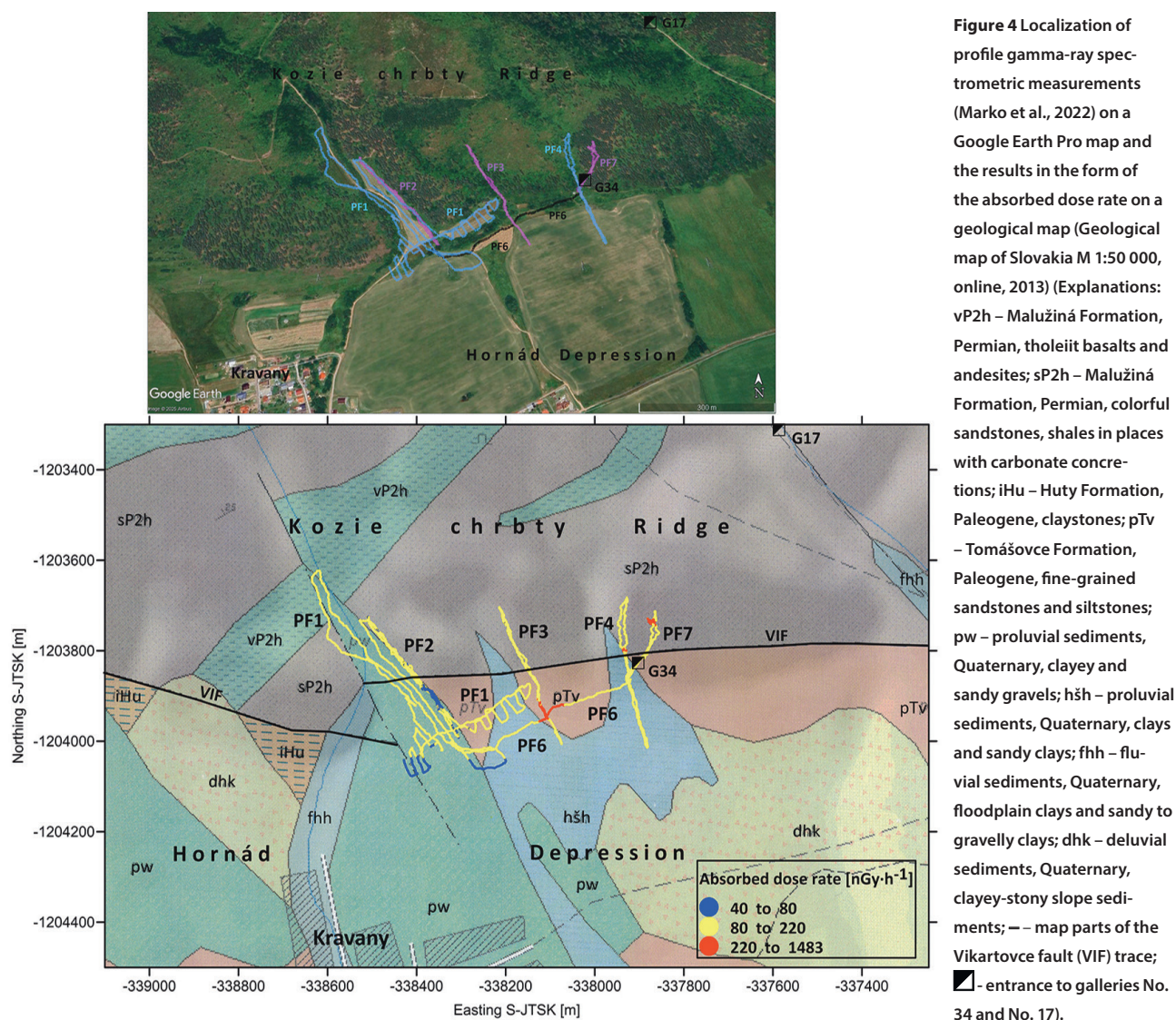
Site	Position	\dot{D}_a [$\text{nGy}\cdot\text{h}^{-1}$]	%K	ppm eU	ppm eTh
PF3	N 49.004337° E 20.206750°	1483	2.95	223.9	23.8
PF4	N 49.005639° E 20.209095°	1300	1.75	191.6	10.2
PF6	N 49.004193° E 20.206848°	694	0.9	109.8	9.9
PF7	N 49.006211° E 20.209838°	613	1.47	98.8	10.7

It follows from a comparison with worldwide (average 2.5 %K, 2 – 4 ppm eU, 8 – 12 ppm eTh by Mareš et al., 1990, and average $55 \text{ nGy}\cdot\text{h}^{-1}$ by IAEA, 1990) or Slovak terrestrial radiation data (average 1.6 %K, 3.3 ppm eU, 9.4 ppm eTh, and $63.3 \text{ nGy}\cdot\text{h}^{-1}$ by Daniel et al., 1996) that the radiation data in Table 1 is very high, especially for the absorbed dose rate \dot{D}_a , even though its spatial range does not look large (Figure 4). It seems that the main reason for such high absorbed dose rates is the uranium concentration, whose maximal values exceed the average of 3.3 ppm eU by several tens of times.

To verify the spatial distribution of these high values, eleven additional survey lines, each approximately 100 meters in length, were projected to carry out gamma-ray spectrometric measurements in rosette shapes with the centers at the PF3, PF4, and PF7 sites (Figure 5). The PF6 site was assigned to the PF3 site, since their maxima positions are very close to each other. The

Table 2 Basic statistical parameters of the measured profiles (red numbers indicate exceeding the average values according to Daniel et al., 1996)

Object	No	\dot{D}_a [nGy·h ⁻¹]				%K				ppm eU				ppm eTh			
		MIN	MAX	AVG	SD	MIN	MAX	AVG	SD	MIN	MAX	AVG	SD	MIN	MAX	AVG	SD
PF1	3820	54.7	129.6	89.9	11.0	0.0	2.3	1.2	0.4	0.2	8.8	2.4	1.1	0.0	14.9	6.4	2.8
PF2	779	94.4	279.8	130.2	31.1	1.0	2.6	1.7	0.3	0.6	31.3	6.5	4.7	2.6	17.6	9.5	2.5
PF3	1085	80.0	1482.6	145.3	205.9	0.4	4.6	1.5	0.4	0.2	223.9	10.3	32.4	1.4	28.1	7.6	2.8
PF4	1002	64.7	1299.8	122.3	160.2	0.4	4.7	1.2	0.4	0.3	191.6	8.0	25.1	1.3	23.7	6.4	2.7
PF6	478	83.0	694.1	178.4	150.7	0.4	3.3	1.4	0.4	0.9	109.8	16.1	24.9	2.4	15.8	7.8	2.5
PF7	869	98.8	613.2	167.0	111.8	0.7	2.8	1.6	0.3	0.7	98.8	13.0	19.6	3.3	16.4	9.5	2.3
PF3 site	694	87.6	1307.3	299.1	280.3	0.6	4.4	1.6	0.5	0.5	194.2	33.7	44.9	3.2	20.7	8.9	3.0
PF4 site	1063	103.4	290.9	127.7	33.7	1.0	2.6	1.7	0.3	0.9	35.0	5.8	5.5	3.7	17.5	9.7	2.3
PF7 site	882	104.8	498.9	178.1	85.1	0.8	3.1	1.8	0.4	0.5	66.8	13.3	14.3	4.4	22.4	11.1	2.7
total	10672	54.7	1482.6	136.4	132.3	0.0	4.7	1.4	0.5	0.2	223.9	8.8	20.1	0.0	28.1	7.9	3.1



statistics of all the measured objects (6 profiles and 3 sites) (Table 2) confirmed again that the average uranium concentrations exceed the Slovak average of 3.3 ppm eU for all objects, except profile PF1 (red color numbers in Table 2). This is valid for the average potassium and thorium concentrations only in the case of profiles PF2 and PF7, and the PF4 and PF7 sites. Finally, the average absorbed dose rates are higher than the Slovak average of 63.3 nGy·h⁻¹ for all objects, and even the minimal absorbed dose rates exceed the average, profile PF1 excepted.

The first interpretation was that these results helped us to confirm the presence of the Vikartovce fault trace as the PF3, PF4 and PF7 sites are close to its map position (Figure 5). But the uranium mineralization in the Kozie chrbtý Mts. is more tied to the lithology (Permian sandstones, conglomerates and shales) than to the faults (Figures 1B and 2). Finding numerous mining pings around the PF7 site on the steep slope turned our attention to the study of old uranium ore mining activities in the area. Tulis & Novotný (1998) reported that, based on geological, geophysical, and borehole works, two mining galleries (No. 34 and No. 17) in the Kravany Beds, with average uranium concentrations 0.05 – 0.4 %, were opened in the Kravany cadaster

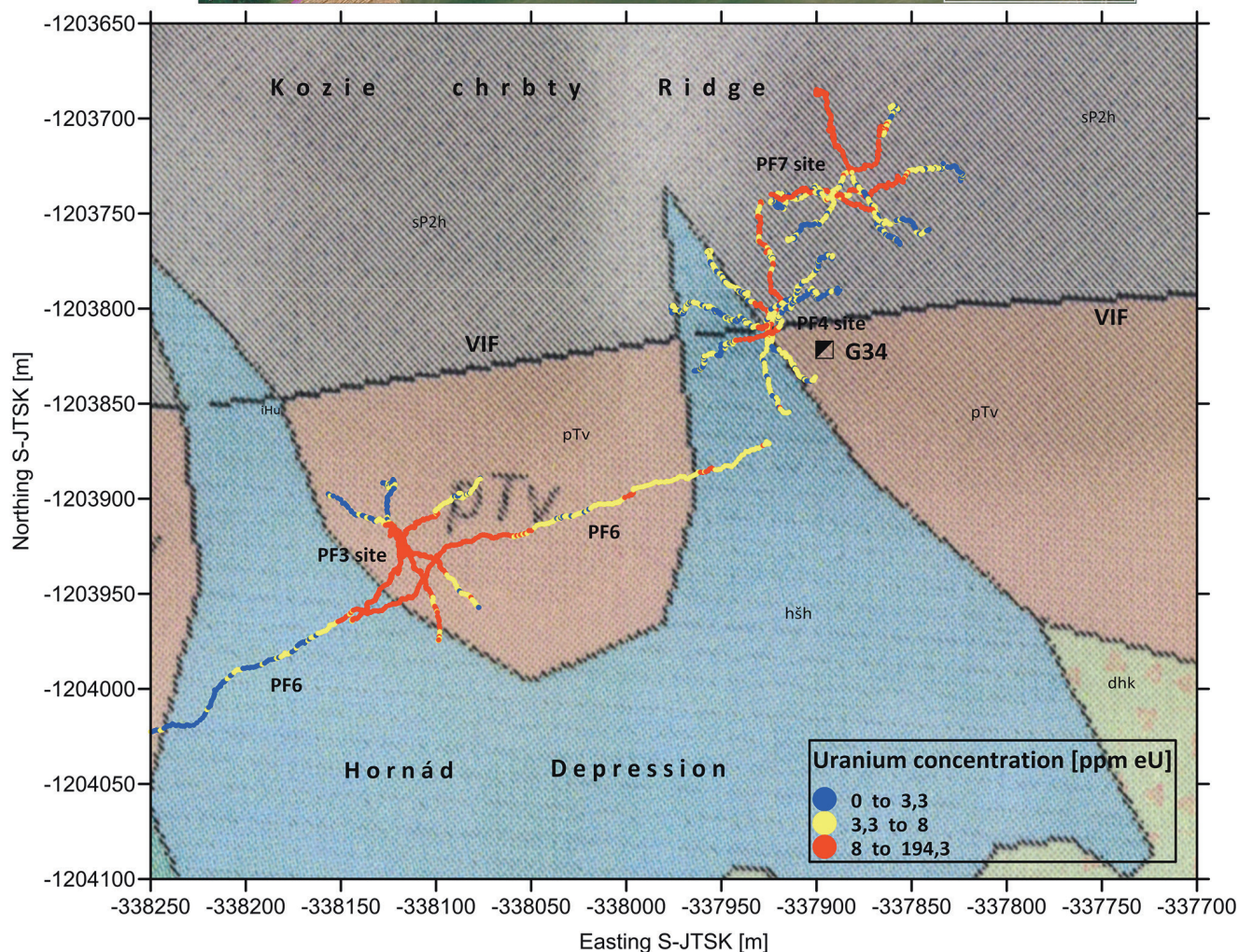
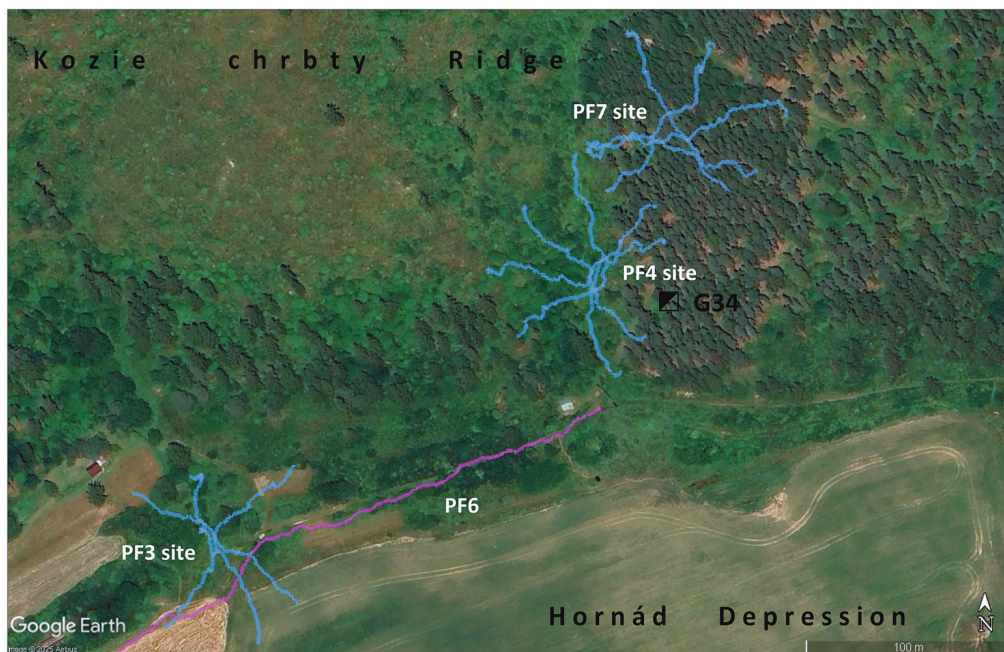
and a total of about 25.5 tons of verified uranium metal were extracted between 1967 and 1972. Both galleries (Figure 4) were closed after exploitation of uranium ore (above 0.03 %) and unprofitability of further mining.

It is evident that the results of our spatial gamma-ray spectrometry measurements reveal the existence of radioactive residues remaining from old uranium ore mining activities. The main mining and operational objects should be entrance into gallery No. 34 close to the PF4 site, mining pings around the PF7 site, the transport field road along the profile PF6, and the handling place at the PF3 site. All these places contain surface radioactive uranium remains that result in increased (above 80 nGy·h⁻¹) and high (above 220 nGy·h⁻¹) levels of natural radioactivity based on the Regulation of the Ministry of Environment of the Slovak Republic No. 1/2000-3 (Figure 6). The area with an increased or a high radioactivity level covers approx. 5.4 ha (hectares) or 5318 m², respectively.

Of course, revised radiometric measurements were carried out at the area after the finishing of uranium mining (Géczyová, 1992). The results of surface radiometric measurements were reported in units of the exposure rate (pA·kg⁻¹; picoampere

Figure 5

Localization of additional profile gamma-ray spectrometric measurements on a Google Earth Pro map and results in the form of the uranium concentration on a geological map (Geological map of Slovakia M 1:50 000, online, 2013) (Explanations as in Figure 4).



per kilogram), where values up to $2.5 \text{ pA}\cdot\text{kg}^{-1}$ represent rocks with normal radioactivity, i.e. radioactively uncontaminated, while values between 2.5 and $3.5 \text{ pA}\cdot\text{kg}^{-1}$ represent increased radioactivity, and values above $3.5 \text{ pA}\cdot\text{kg}^{-1}$ represent anomalous

radioactivity; both higher levels indicate a radioactively contaminated environment (Tulis & Novotný, 1998). Using the IAEA (2003) conversion coefficient between the exposure rate and the absorbed dose rate ($1 \text{ pA}\cdot\text{kg}^{-1} = 121.19944 \text{ nGy}\cdot\text{h}^{-1}$), Figure

7 shows the spatial distribution of the uncontaminated and contaminated ground environment in the study area, where $2.5 \text{ pA}\cdot\text{kg}^{-1} \approx 303 \text{ nGy}\cdot\text{h}^{-1}$ and $3.5 \text{ pA}\cdot\text{kg}^{-1} \approx 424 \text{ nGy}\cdot\text{h}^{-1}$. In this case, the area coverage of the radioactively contaminated environment is restricted mostly to the PF3 site, where the increased (above $303 \text{ nGy}\cdot\text{h}^{-1}$) radioactivity level covers approx. 1912 m^2 , including approx. 953 m^2 of the anomalous (above $424 \text{ nGy}\cdot\text{h}^{-1}$) radioactivity level.

To evaluate the health risk for humans that arises from terrestrial radioactivity in the study area, an assumption of a stay out-of-doors all year round needs to be considered. World estimates of average outdoor absorbed dose rate in the air $55 \text{ nGy}\cdot\text{h}^{-1}$ - give an annual effective dose equivalent of 0.34 mSv (milisievert) (IAEA, 1990). The whole extrapolated studied area above $80 \text{ nGy}\cdot\text{h}^{-1}$ in Figures 6 and 7 thus represents a place with

a higher-than-average health risk, with an annual effective dose equivalent above 0.5 mSv . A closer overview of area coverage with higher levels of natural radioactivity is presented in Table 3.

Table 3 The extent of surface coverage [m²] by the higher levels of natural radioactivity of the extrapolated study area.

Object	Absorbed dose rate \dot{D}_a [nGy·h ⁻¹]			
	> 80	> 220	> 303	> 424
PF3 site		3166	1912	953
PF4 site		118	0	0
PF7 site		2034	148	0
total	54428	5318	2060	953
	> 0.5	> 1.36	> 1.87	> 2.62
Effective dose equivalent E [mSv·a ⁻¹]				

Figure 6 Localization of additional profile gamma-ray spectrometric measurements on a Google Earth Pro map and results in the form of natural radioactivity levels (Regulation No. 1/2000-3) on a geological map (Geological map of Slovakia M 1:50 000, online, 2013) (Explanations as in Figure 4).

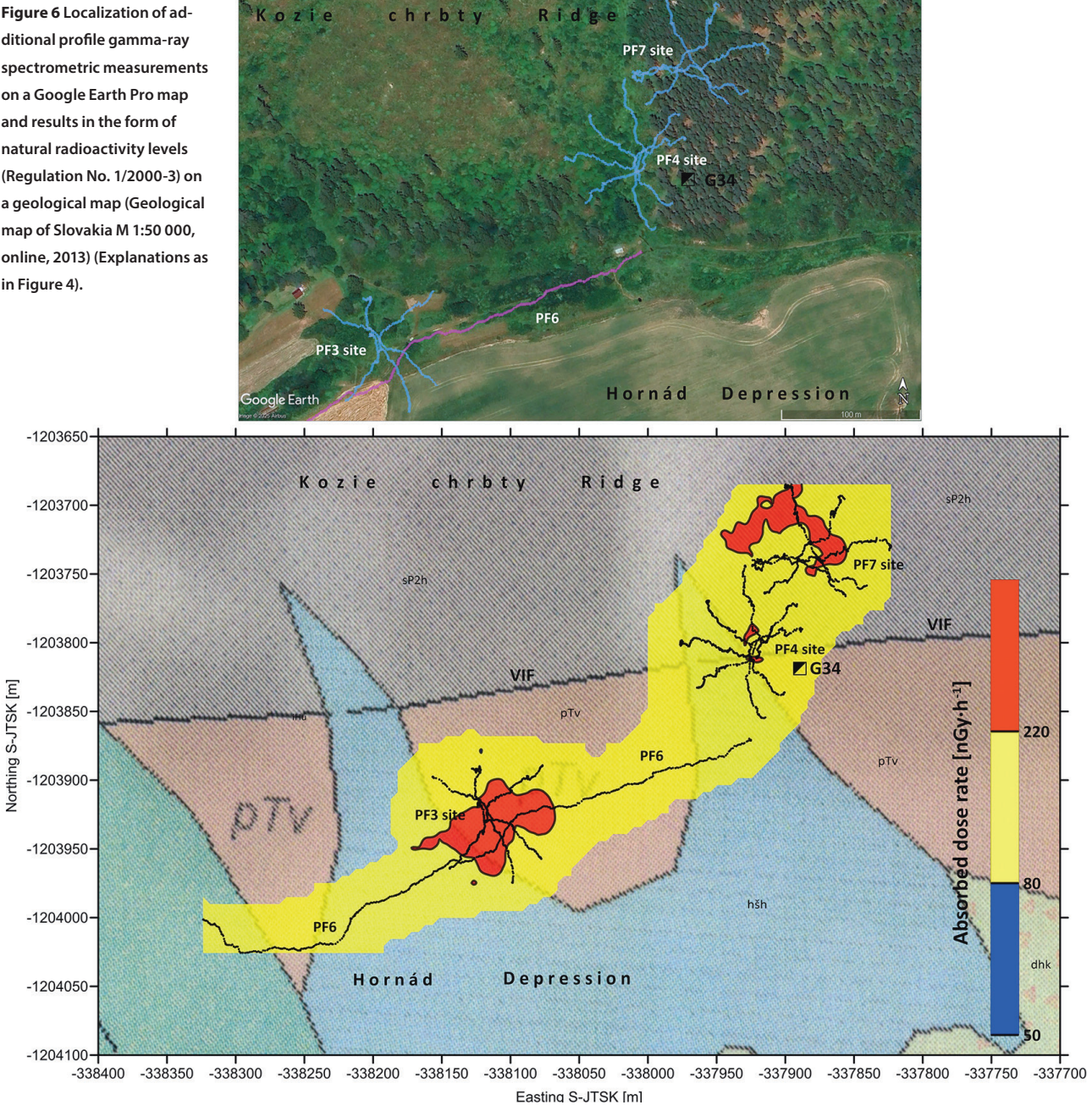
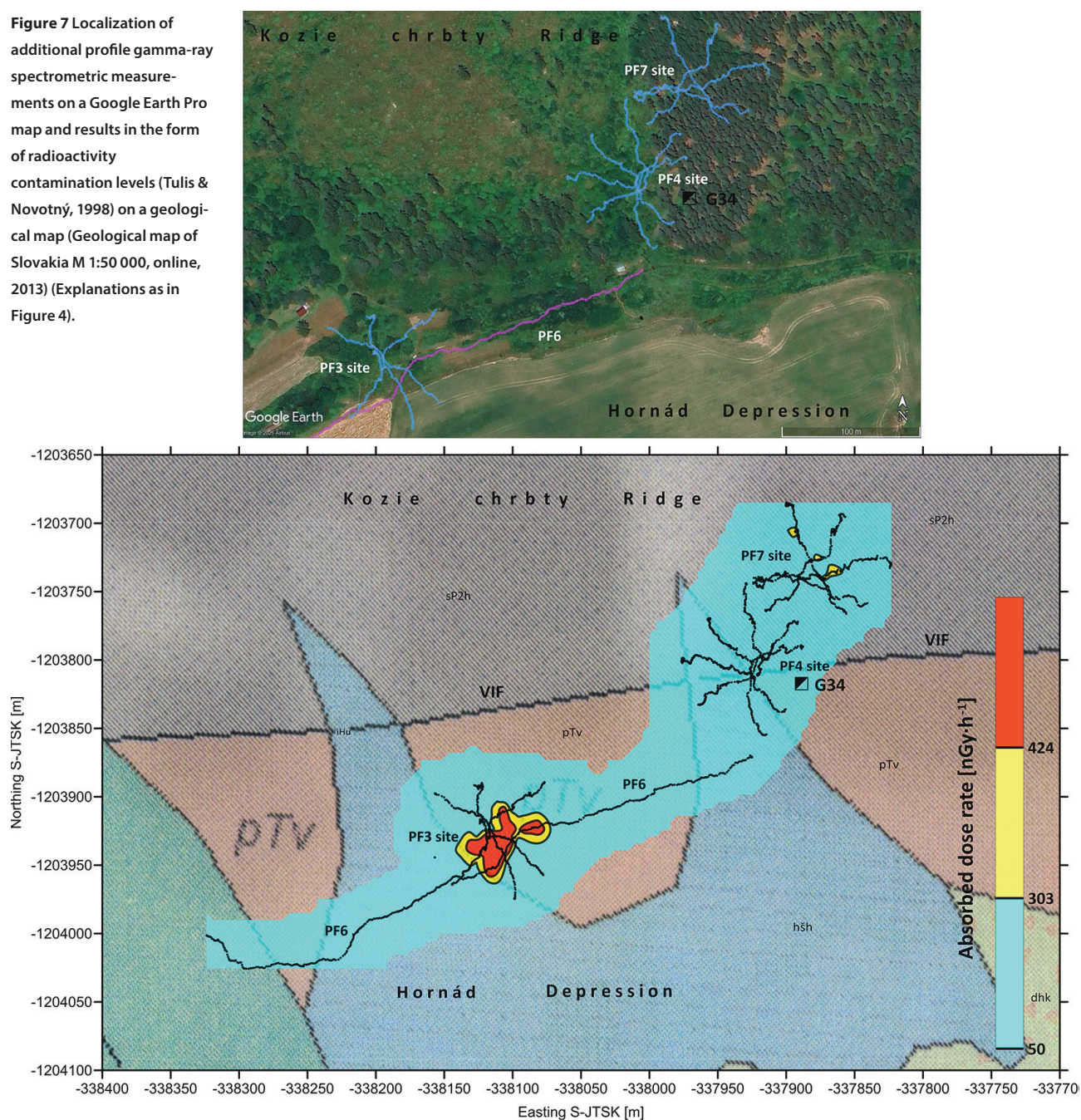


Figure 7 Localization of additional profile gamma-ray spectrometric measurements on a Google Earth Pro map and results in the form of radioactivity contamination levels (Tulis & Novotný, 1998) on a geological map (Geological map of Slovakia M 1:50 000, online, 2013) (Explanations as in Figure 4).



CONCLUSIONS

The increased and high values of terrestrial radioactivity measured and confirmed by the profile ground gamma-ray spectrometry method in the north-eastern vicinity of the village of Kravany have their origin in near-surface contamination by the remnants of uranium ore mining and handling at several places. Namely, the uranium concentration reaches the maximal values up to 223.9 ppm eU, and the maxima of the absorbed dose rate subsequently reach values up to 1482.6 nGy·h⁻¹, which exceed the average values on the order of 10- to 100-times.

These conclusions agree with the formulations in the Tulis & Novotný (1998) report: “Despite the fact that the liquidation was carried out on the basis of approved liquidation plans and with

the aim of minimizing the impact on the ecology of the region, it cannot be ruled out that in the course of the next 20 – 30 years certain sections or parts of the deposits may be found that will not comply with the currently valid legislation in terms of environmental protection. This is also indicated by the information from pp. 126 – 128 (taken from the report by Géczyová, 1992), where in several places exposure rate values higher than 3.5 pA·kg⁻¹ are found, which from the point of view of the valid standards indicate a polluted area. These are the deposits of Spišský Štiavnik, Švábovce, Kravany and Vikartovce, where the maximum measured exposure rate values reached 20 – 63.9 pA·kg⁻¹. The above facts point to the usefulness of further review works documenting the impact of U-mineralization on individual components of the environment (air, soil, water, rock environment)”.

It should also be mentioned here that no visible remains of mining works, structures, and equipment have been registered in the study area, except for several pings on the steep slope at the PF7 site. In view of the relatively easy accessibility of the PF6 profile and PF3 site areas, it is appropriate to warn against the use of these sites for human activities, especially the construction of residential or farm buildings that would mean a longer stay of people (some buildings are already identifiable in Google Earth maps). Reclamation of the rock and soil environment or other measures would be necessary in these cases.

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