Slovak Geol. Mag., 3 (1997) 67–77

Geochemical Atlas of Slovak Republic Part Natural Radioactivity

JOZEF DANIEL[1], LADISLAV LUČIVJANSKÝ[1], MARIÁN STERCZ[1], SLAVOMÍR DANIEL[2]

[1]URANPRES, Ltd., Fraňa Kráľa 2, 052 80 Spišská Nová Ves
[2]Geological Survey of Slovak Republic, regional centre, Markušovská cesta 1, 052 40 Spišská Nová Ves

Abstract. The Atlas of Natural Radioactivity of Slovakia is one of the several separate parts of the Geochemical Atlas of the Slovak Republic prepared at a scale 1 : 1 000 000. The Atlas evaluates natural radioactivity of rocks and waters. Assessed are K, U, Th, U_{total} components and dose rate of natural radioactivity of rocks, Rn on radon-risk maps, as well as U_{nat}, ^{226}Ra and ^{222}Rn components of natural radioactivity of waters. Aside from these maps, the Atlas also includes an assessment of cosmic radiation dose rate.

Key words: Natural radioactivity, rocks, waters, cosmic radiation, Slovakia.

Introduction

The investigations of radioactivity in the natural environment were closely related to the intensive prospecting and exploration of uranium ores after World War II. Geological exploration and mining of radioactive minerals in Slovakia persisted, with short breaks, from 1947 to 1990. They were carried out by a revision detachment of the company Jáchymov Mines, and, since 1960, by the separate Slovak Division of the Uranový pruskum (Uranium Survey). The exploration of radioactive minerals comprised various radiometric techniques (airborne gamma survey, car-borne gamma survey, surface radiometry - mountain-topography measurements, surface gamma survey, gamma survey in pits, gamma spectrometry, emanation survey) and instruments of different kinds, purpose and accuracy.

The surveys were distributed over all of the West Carpathian geological units, but were mainly focussed on the crystalline units of the core mountains, Permian formations, Neogene volcanic mountain ranges and Tertiary basins.

One of the objectives of the project Investigations of Geological Factors of Environment carried out by the Dionýz Štúr Institute of Geology in association with other organizations between 1990 and 1995 was to compile natural-radioactivity maps at a scale of 1 : 200 000 and in some areas of Slovakia at 1: 50 000. The works aimed at radioactivity were performed by URANPRES, Ltd. based at Spišská Nová Ves, Geocomplex, Inc., based in Bratislava, and C & S Radón, Ltd. based at Spišská Nová Ves.

All these works were used to compile the Atlas of Natural Radioactivity of Slovakia at scale 1 : 1 000 000 (Daniel, Lučivjanský & Stercz, 1996).

Natural Radioactivity and Its Measurements

The maps of the natural radioactivity of rocks are based on gamma spectrometric measurements. The investigations were made at a scale of 1 : 200 000 covering Slovakia on a uniform grid 3 x 3 km, i. e. 1 point per 9 km^2. Six selected areas (Upper Nitra Valley, Nízke Tatry Mts., Hornád Basin and the eastern part of the Slovenské rudohorie Mts., Košice Basin and Slánske vrchy Mts., Žiar Basin and Malá Fatra Mts.), which account for 20 % of Slovakia's territory were investigated at scale 1 : 50 000 on a uniform grid 1 x 1 km, i. e. 1 point per 1 km^2. The measurements were taken by a reference-point technique whereby 1 reference point represents 5 cross-like distributed spectrometric measurements located 20 m from the middle of the cross. The average value of these 5 readings corresponds to the value of the relevant reference spectrometric point. The measure-

Tab. 1 Categories of foundation-soil permeability

Categories of foundation-soil permeability	Earth categories acc. to state standard 73 1001
poorly permeable f > 65 %	F5, F6, F7, F8
moderately permeable 15 % < f < 65%	F1, F2, F3, F4, S4, S5, G4, G5
strongly permeable f < 15 %	S1, S2, S3, G1, G2, G3

ments were taken by GS-256 instruments, which were regularly calibrated and checked. Check measurements taken near the border between Slovakia and the Czech Republic revealed that measurements in both countries are highly compatible.

The rock radiometric maps are based on the results of gamma spectrometry. The measurements were taken on a total of 15 573 reference spectrometric points, corresponding to 77 865 gamma-spectrometric measurements points.

The radon-risk derivative map at a scale of 1 : 200 000 was made by the SAN technique designed by the company Uranium Survey, Liberec. The measurement sensitivity was 1 kBq . m^{-3}. More up-to-date measurement techniques, mainly Radon detection through Lucas chambers, are currently being used. The main characteristics to assess radon risk comprise ^{222}Rn volume activity given in kBq . m^{-3} and foundation-soil category (Slovak standard 73 1001) from which gas-permeability of rocks is derived.

The main assessment characteristics are given in Tables 1 and 2.

Tab. 2 Categories of radon risk

Radon risk	^{222}Rn volume activity in soil air (kBq . m^{-3}) in foundation soils according to gas permeability		
	Gas permeability		
	low	medium	high
low	<30	<20	<10
medium	30-100	20-70	10-30
high	>100	>70	>30

The natural radioactivity of waters was investigated by field collecting and laboratory measurements of water samples. The samples were taken from springs, wells, drillholes, outflows from mine workings, rivers, lakes and tailing ponds. The sampling density was 1 sample per 10 km^2 at a scale of 1 : 200 000 and 1 sample per 5 km^2 at a scale of 1 : 50 000.

Natural Radioactivity Maps of Slovakia

The natural radioactivity maps of Slovakia illustrate the distribution of sources of natural radioactivity of rocks, water radioactivity, radon risks and cosmic radiation in Slovakia.

Cosmic Radiation Dose Rate Map (Da_{cosmic})

The cosmic radiation dose rate map was based on C. Murith's and A. Gurtner's (1990-93) investigations. Cosmic radiation dose rate depends on the altitude of the territory. The lowest place in Slovakia near Streda nad Bodrogom (94 m above sea level) has an average cosmic radiation dose rate 38.3 nGy . hr^{-1}. The highest spot is 2 655-m-high Mt. Gerlachovský štít whose cosmic dose rate amounts to 101.5 nGy . hr^{-1}.

Potassium Concentration Map

Potassium is a rock-forming element whose average abundance in the earth's crust is 2.52 weight %. Its abundance in particular rock types are as follows (Polánski & Smulikowski, 1971):

– igneous rocks :	– ultramafic	0.03 %
	– mafic	0.83 %
	– acid	3.34 %
– sedimentary rocks:	– argillaceous	2.66 %
	– arenaceous	1.07 %

Of the three potassium isotopes, only ^{40}K is radioactive.

The potassium concentration map of Slovakia (Fig. 1) reveals that potassium values range from extremely low 0.1 % in Mesozoic limestones and dolomites in the Malá Fatra Mts. to 5.4 % in Lower Triassic rocks of the Melaphyre series.

The average value calculated from all masurements taken in Slovakia amounts to 1.66 %, which is less than the average content in the earth's crust (2.6 %).

Thorium Concentration Map

Of six naturally occurring thorium isotopes, only ^{232}Th (parent element of the thorium series) is significantly radioactive.

The average thorium content in the earth's crust is usually put at 8–12 ppm. It is 18 ppm in acid igneous rocks, 3 ppm in mafic rocks, 11 ppm in shales and clays, 10 ppm in sandstones and 1.8 ppm in limestones.

The average concentration in Slovakia is 9.4 ppm. The measured values range greatly from a few ppm to as much as 26 ppm.

The lowest thorium contents of 6.8 ppm (Fig. 2) occur in the Záhorie – Lower Moravian sector of the Vienna Basin. The contents in the Malé Karpaty Mts. vary from 5 to 12 ppm and in the Central Slovak Neogene Volcanic Mountains they average 9.7 ppm Th. The highest values (21.2 ppm) are found in Neogene rhyolites. Thorium contents amount to 9 ppm in the Nízke Tatry and 9.6 ppm in the eastern part of the Slovenské rudohorie Mts. Low thorium contents occur in the Slovak Paradise (7.9 ppm) and Slovak Karst (10 ppm).

Uranium Concentration Map

The average U content in the earth's crust is put at 2.3 ppm. In igneous rocks the content grows with increasing acidity from 0.5 ppm in mafic rocks to 1.8 ppm in intermediate rocks and to 3.5 ppm in acid igneous rocks (Matolín M., 1994). The average content in shales and clays is 4.0 ppm, in sandstones 3.0 ppm and in limestones only 1.4 ppm U. The mean value is 2.9 ppm (Matolín M., 1976).

Gammaspectrometric measurements yielded an average value of 3.3 ppm, the highest contents exceeding 10 ppm.

Concentrations of uranium minerals form uranium deposits in some places.

Major deposits in Slovakia include those in the Spišská Nová Ves (Novoveská Huta) - Hnilčík - Malý Muráň area, in the Hronicum of the Nízke Tatry, and in the Považský Inovec Mts. (Kálnica, Selec).

The Košice deposit near the tourist resort of Jahodná may be developed in the future.

The lowest U contents of 2.5 ppm are found in the Vienna Basin (windblown sands). The biggest variations occur in the Central Slovak Neogene Volcanic Mountains (0.6–16 ppm eU). Uranium concentrations average 3.1 ppm in the Slovenské rudohorie, 3.3 ppm in the Eastern Slovak Neogene Volcanic Mountains and 3.8 ppm in intramontane basins (Fig. 3).

Map of Gamma Radiation Dose Rate of Rocks

The map of gamma radiation dose rate was compiled from adjusted relationships between the contents of U, Th, K and dose rate (Fig. 4). A look at the dose-rate map reveals that the lowest values occur along the Slovak borders, whereas the highest ones are found in central Slovakia.

The average gamma radiation dose rate value in Slovakia is 63.3 nGy . hr^{-1}.

In most countries, radioactivity measurements are plotted on a map of rock dose rates. The values measured in Slovakia can be compared with an updated table of average dose rate values in some other countries for which the data were available (Tab. 3).

Tab. 3 Average dose rate values in some other countries

Country	Dose rate Ngy . hr^{-1}		Number of measurements	Year of compilation	Measurement technique
	average	range			
Australia	43	20–150	> 1 000	1980	ground
Belgium	43	13–58	272	1987	ground
Czech Republic	66	6–245	100 % of area	1995	airborne
Denmark	38	17–52	14 areas	1980	ground
Eire	42	0–180	264	1980	ground
Finland	65			1980	
France	68	10–250	5 142	1985	ground
Germany (East)	85	24–270	1 005	1969	ground
Germany (West)	53	4–350	24 739	1978	ground
Great Britain	40	0–100	1 400	1984	ground
Hungary	55	20–130	123 areas	1987	ground
India	55	20–1100	2 800	1986	ground
Island	28	11–83		1982	
Italy	57	7–500	1 365	1972	ground
Japan	49	5–100	1 127	1980	ground
Canada	24	18–44	33 areas	1984	airborne
Netherlands	32	10–65	1 049	1985	ground
Norway	73	20–1200	234	1977	ground
Poland	34	10–110	19 528	1994	ground
Romania	81	32–210	2 372	1979	laboratory
Slovakia	63	3–179	15 573	1996	ground
Slovenia	57	4–140	1 052	1993	ground
Sweden	80	18–4000		1979	
Switzerland	48	5–368	805	1995	ground airborne
Taiwan	69		26	1972	laboratory
USA	46	13–100	25 areas	1972	airborne

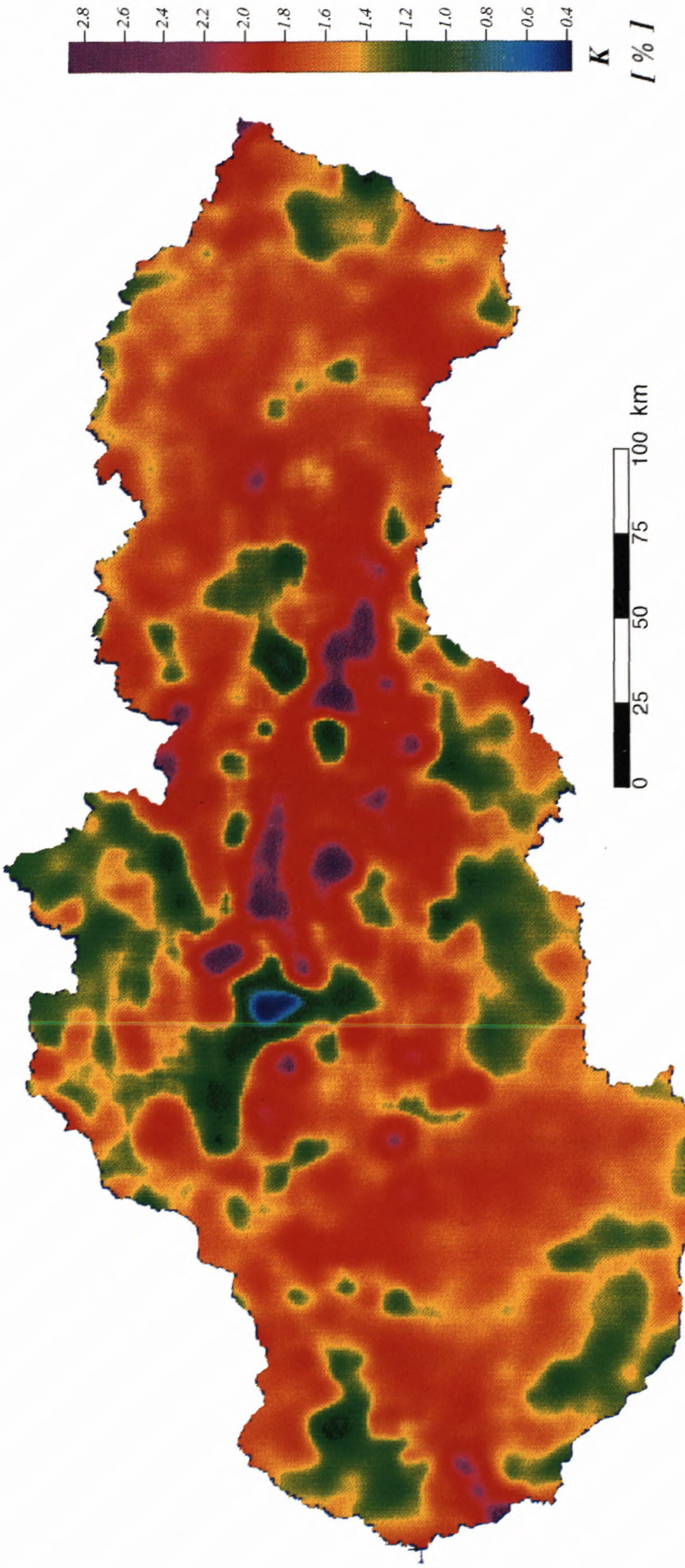

Fig. 1 Map of potassium concentration

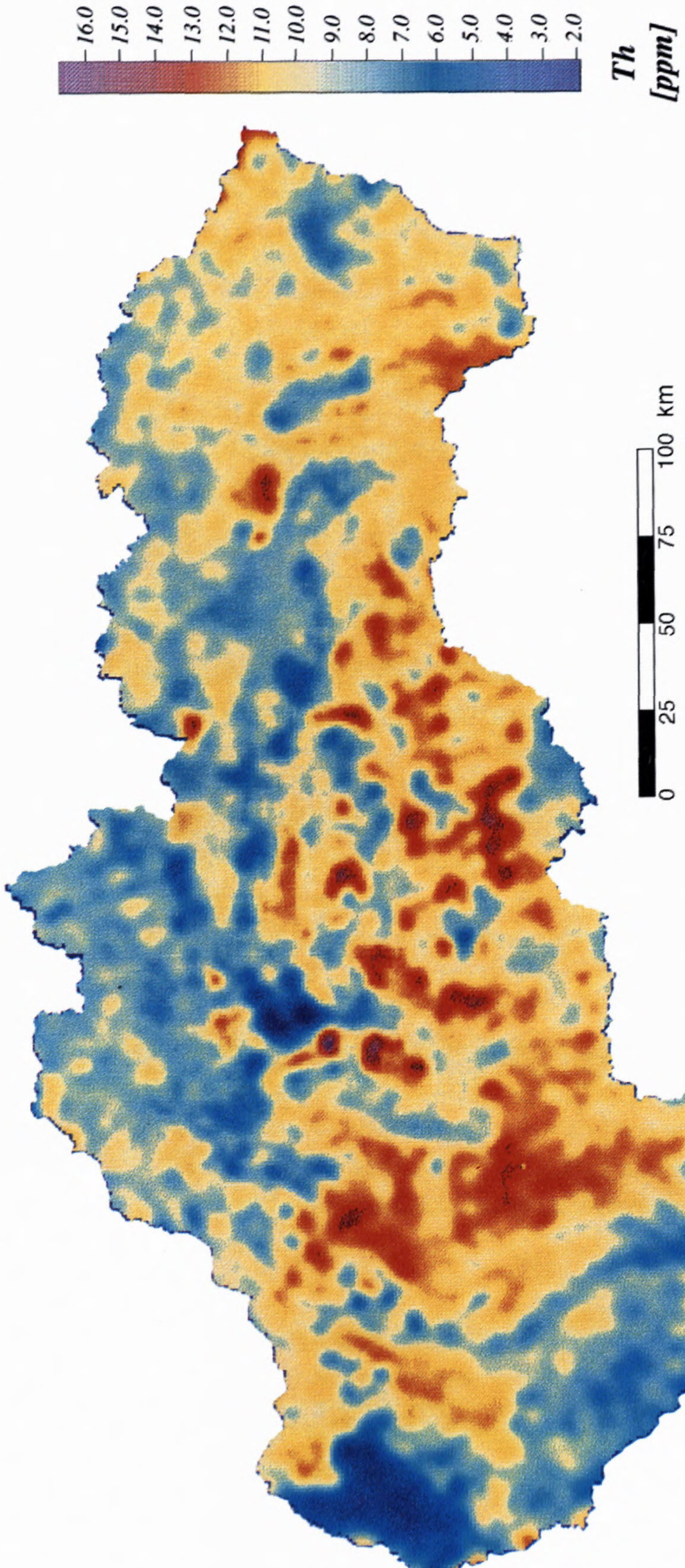

Fig. 2 Map of thorium concentration

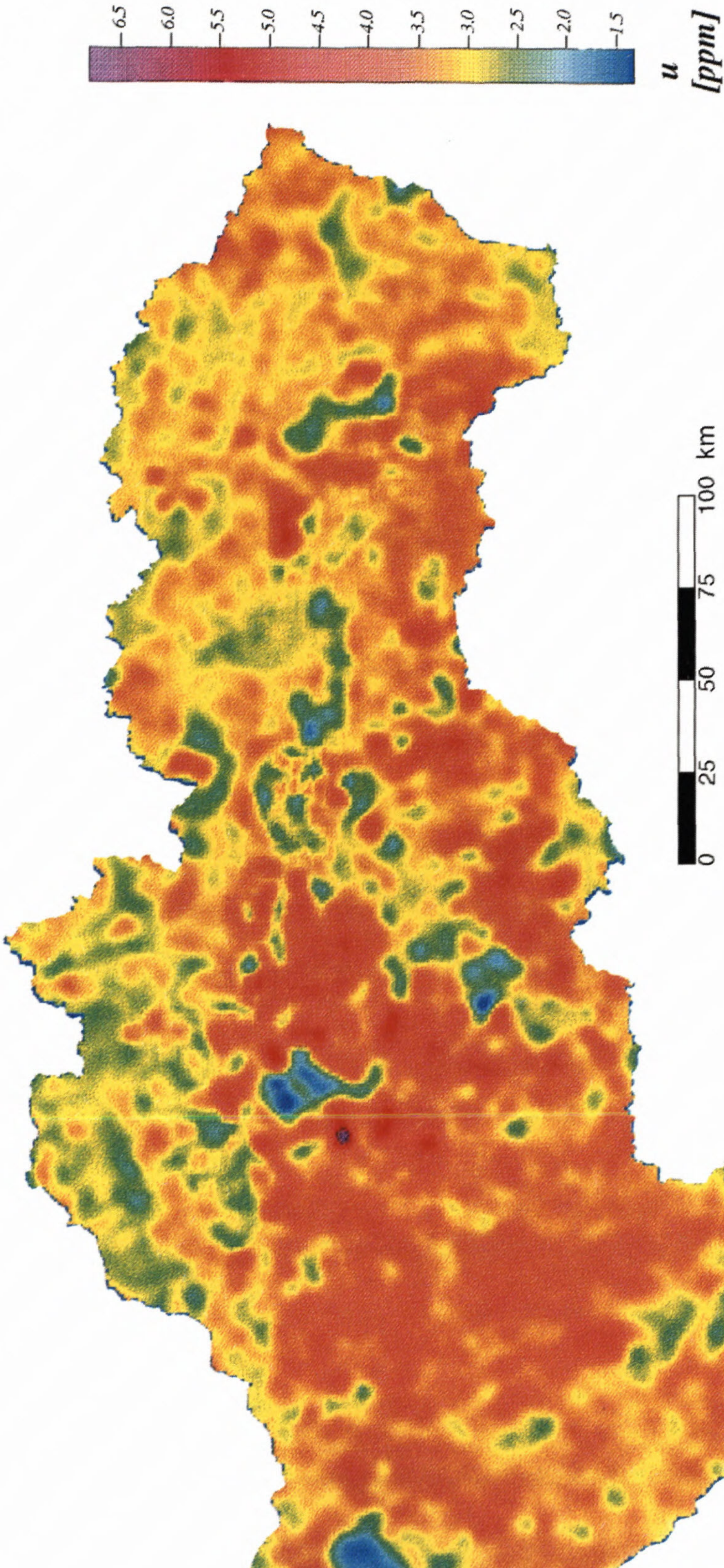

Fig. 3 Map of uranium concentration

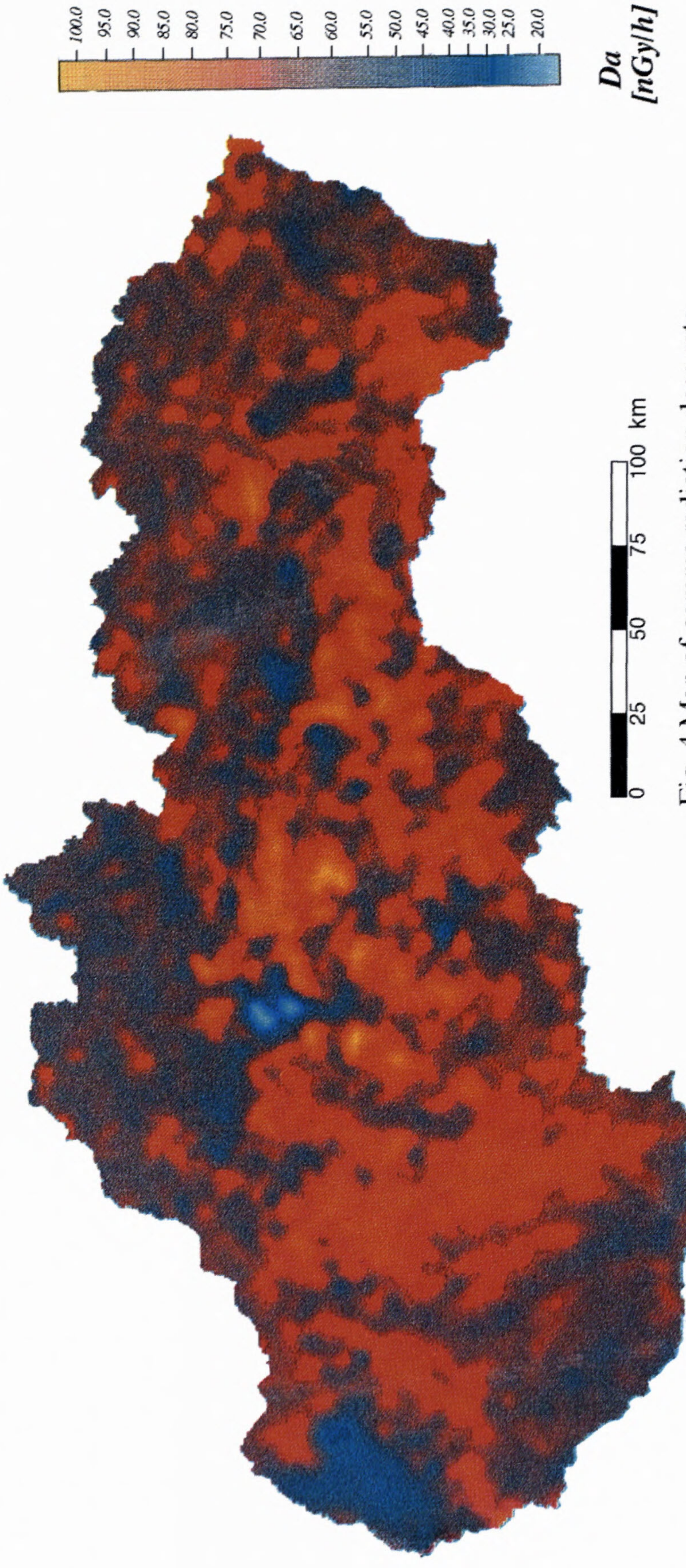

Fig. 4 Map of gamma radiation dose rate

Derived Map of Radon Risk

The derived map of radon risk (Fig. 5) was compiled from measured data and all archive data on natural radioactivity (Čížek P., Smolárová H., 1993).

Slovakia's territory was divided into three categories of radon risk. The shares of respective categories on Slovakia's territory are as follows:

– low radon risk 53.0 %
– medium radon risk 46.7 %
– high radon risk 0.3 %

The highest radon risk in areas measured at a scale of 1 : 50 000 occurs in the area of the town of Levice (15.7 % was in the low, 75.6 % in the medium and 8.7 % in the high radon risk category).

As for geological units, the highest values are found in the Gelnica and Rakovec Groups of the Gemericum in the Slovenské rudohorie. The Middle and Upper Triassic dolomites, called uranium dolomites, are noteworthy in this respect, too.

Maps of Natural Radioactivity of Waters

A total of 5,271 water samples were collected in Slovakia's territory and were used to compile the maps of natural radioactivity of waters.

Map of Uranium Concentration in Waters

Increased uranium concentrations in groundwaters occur largely in waters draining uranium exploration and exploitation mine working (Novoveská Huta, Kálnica), waters in the vicinity of uranium occurrences (Východná) and in some thermal waters (Oravice, Lúčka by Spišské Podhradie). Increased concentrations are also present in waters of some core mountains underlain by the crystalline unit (Považský Inovec, Malá Fatra), in dolomites of the Choč nappe (Nízke Tatry) and in the Danube Lowland and Southern Slovak Basin. The average U_{nat} content in Slovakia's waters amounts to 0.003 mg . l^{-1}.

Map of Radium Concentration in Waters

The ^{226}Ra volume activity ranges from 0.001 to as much as 9.7 Bq . l^{-1}. The highest values are characteristic of mineral and thermal waters. Increased values also occur in waters of the Záhorie Lowland and western part of the Danube Lowland, in some core mountains (Malé Karpaty, Považský Inovec), Hornád and Poprad Basins, Galmus and the western part of the Volovské vrchy Mts. In Paleogene rocks, radium volume activity grows with increasing depth of groundwater circulation. Radium contents in waters of Neogene volcanics in the Slánske vrchy and Vihorlat Mts. are twice as high as those in waters of Central Slovak Neogene Volcanic Mountains.

Map of Radon Concentration in Waters (Fig. 6)

Radon concentration in water depends on the presence of rocks with uranium minerals, tectonics, water temperature and T. D. S. According to Lange's (1969) classification, several kinds of radon waters can be distinguished.

The first type of radon waters present in nearly all core mountains is radon water formed in fractured weathering crust of crystalline units in core mountains. Such waters are most widespread in the crystalline units of the Veporské vrchy and Stolické vrchy Mts.

Another type consists of radon water bound to clayey-travertine deposits. It includes mineral waters at Sivá Brada near Spišské Podhradie and Bešeňová.

A third type comprises radon waters bound to deep faults, characterized by big discharges and increased temperatures. This type is exemplified by the mineral spring "Carbonic" at Oravice.

Radon water bound to faults is fairly widespread. The type comprises water in the vicinity of the Subtatric fault (Žiar, Starý Smokovec), in the Košice and Hornád Basins.

A fifth genetic type consists of water draining the uranium deposits and springs located near uranium occurrences (Novoveská Huta near Spišská Nová Ves, Kálnica, Východná).

Radioactivity of Mineral and Thermal Waters in Slovakia

In comparison with normal waters, cold mineral waters are enriched in radionuclides, particularly radium and radon. In some thermal waters, radium volume activity is a hundred times as high as in normal waters.

Investigations of natural radioactivity of more than 200 samples of Slovakia's mineral waters allowed us to divide the mineral waters into several categories:

– mineral water bound to Triassic carbonates, mainly those of the Krížna and Choč nappes. Uranium concentrations in these water are low, averaging less than 0.003 mg . l^{-1}. Radium volume activity mostly ranges from 0.2 to 0.9 Bq . l^{-1}. Radon volume activity largely varies between 20 and 50 Bq . l^{-1} but in some water it attains even higher values. Springs of these waters are frequently coated with travertine.

– mineral water of the crystalline unit is mostly cold acidic. They are enriched in uranium (0.005–0.015 mg . l^{-1}), radium (^{226}Ra volume activity largely varies from 0.1 to 0.5 Bq . l^{-1}) but mainly radon (often over 200 Bq . l^{-1}).

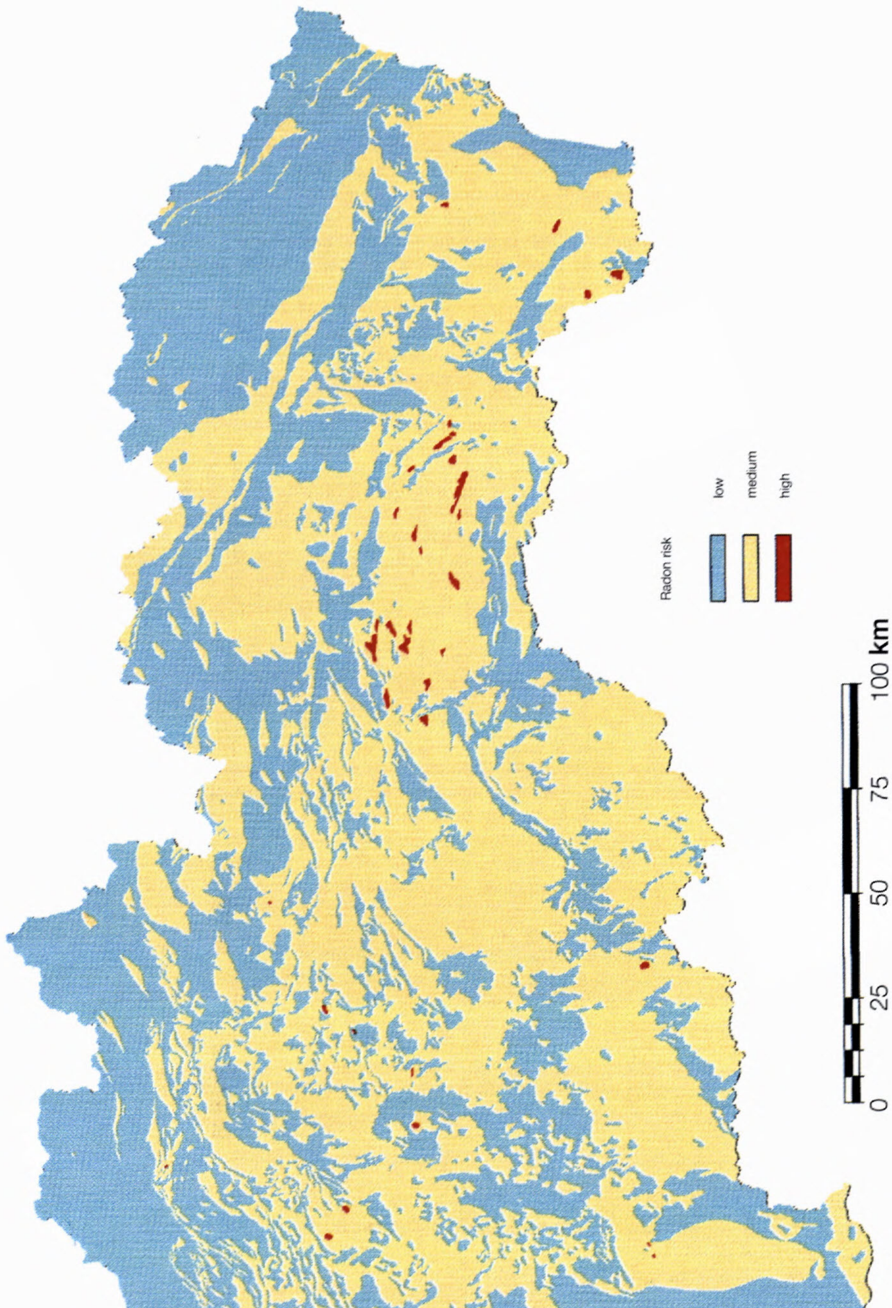

Fig. 5 Derivative map of radon risk

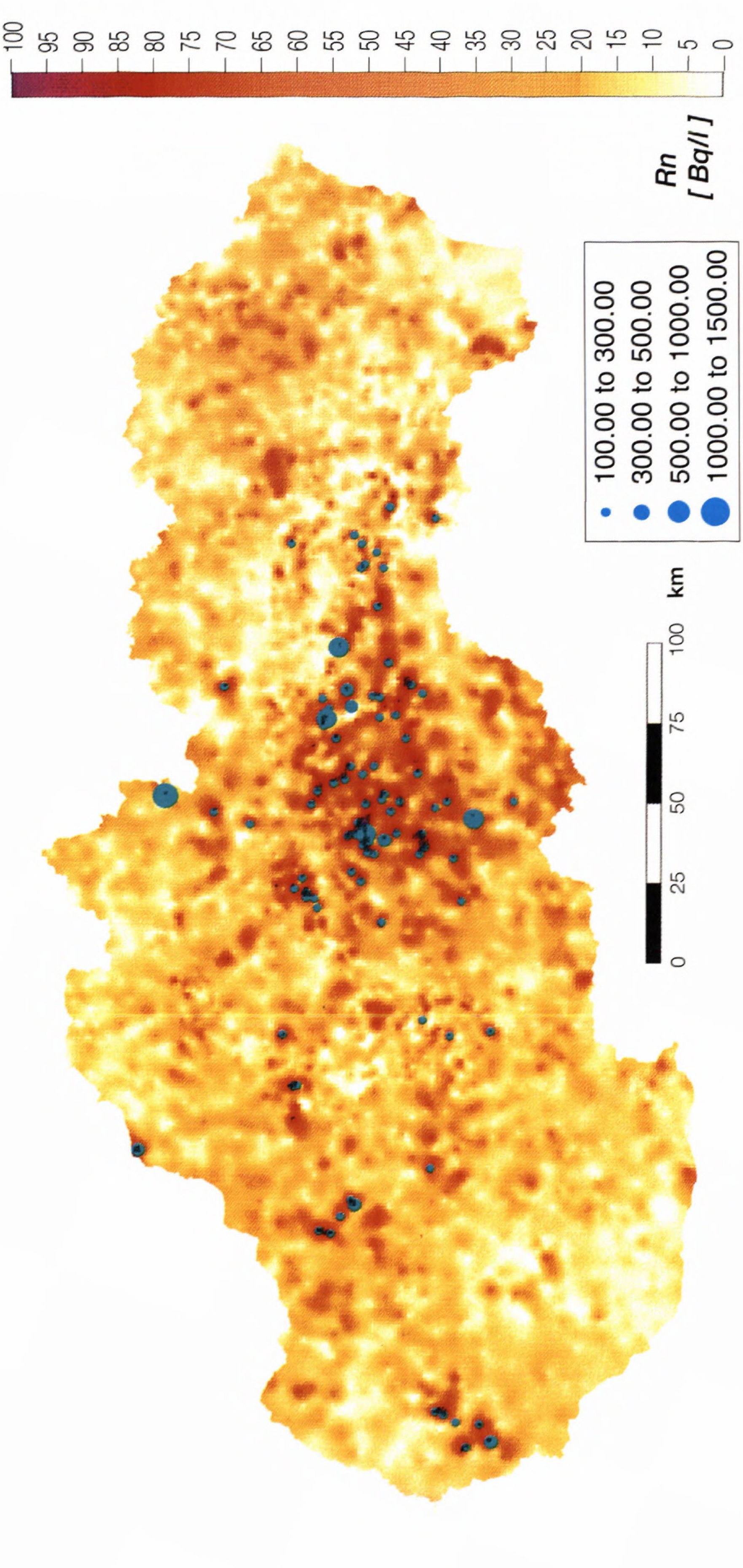

Fig. 6 Map of radon concentration in water

– mineral water of the Flysch Belt - sodium-bicarbonate hydrogensulphide or sodium-bicarbonate acidic water. Water bound to tectonic lines occurs in all partial units of the Flysch Belt. It is typically slightly enriched in uranium (0.004 Bq . l^{-1}), their ^{226}Ra volume activity being medium (0.02–0.08 Bq . l^{-1} only rarely over 0.1 Bq . l^{-1}). ^{222}Rn volume activity varies from 10 to 20 Bq . l^{-1}, only in water enriched in CO_2 does it exceed 20 Bq . l^{-1}.

– mineral water of Neogene volcanic mountains – it is characterized mostly by low uranium (0.002 mg . l^{-1}) and radium contents (up to 0.05 Bq . l^{-1}) and by mildly increased ^{222}Rn volume activities (20–40 Bq . l^{-1}).

With regard to their natural radioactivity, Slovakia's natural water falls into two major categories:

– thermal water of pre-Tertiary units – characterized by high ^{226}Ra volume activity. The category includes water at Bešeňová, Piešťany, Oravice (well OZ-2), Lúčka (well BŠ-1), Vrbov, Kováčová, Lúčky, Trenčianske Teplice, etc. It is largely associated with Triassic rocks of the Krížna and Choč nappes.

– thermal waters of Tertiaty units – characterized by low ^{226}Ra volume activity (up to 0.1 Bq. l^{-1}). This category is exemplified by the thermal waters at Dunajský Klátov, Vlčany, Baloň, Tvrdošovce, Sládkovičovo, Diakovce, Nové Zámky and Topoľníky.

Conclusions

The Atlas of Natural Radioactivity of Rocks was compiled from a multitude of measurements, results and reports. Most data resulted from in-situ field measurements at scales of 1 : 200 000 and 1 : 50 000. Results of works performed by URANPRESS Ltd. Spišská Nová Ves during more than 35 years of exploration for radioactive minerals, the Geological Survey of the Slovak Republic of Bratislava, the C & S Radón Ltd. of Spišská Nová Ves and the Geocomplex of Bratislava were used as well. A further major source of information and instruction was M.Matolín's publication "Radioactivity of West Carpathian Rocks" (1976) which first summarized work on radioactivity in Slovakia. It resulted also in the first map of total radioactivity at a scale of 1 : 500 000.

Natural radioactivity in Slovakia can be characterized by the following average values:

Natural radioactivity of rocks:

Potassium (K)	1.6 %
Thorium (eTh)	9.4 ppm
Uranium (eU)	3.3 ppm
Dose rate (Da)	63.3 nGy . hr^{-1}

Radon risk:

– low	53.0 %
– medium	46.7 %
– high	0.3 %

Natural radioactivity of waters:

Uranium (U_{nat})	0.003 mg . l^{-1}
Radium (Ra)	0.040 Bq . l^{-1}
Radon (Rn)	9.31 Bq . l^{-1}
Cosmic radiation	
Dose rate (Da)	44.3 nGy . hr^{-1}

The natural radioactivity maps are considered to be environmental maps, inasmuch as they can be used to assess human irradiation.

References

Čížek, P. & Smolárová, H., 1992: Derivated maps of Radon risk of Slovakia at scales 1: 200 000 (in Slovak). Manuscript Uranpres, Spišská Nová Ves.

Daniel, J., Lučivjanský, L. & Stercz, M., 1997: Geochemical Atlas of Slovak Republic, part Natural Radioactivity, in press

Lange, O., K., 1969: Hydrogeology (in Russian). "Vyššaja škola", Moscow.

Matolín, M., 1976: Radioactivity of West Carpathian Rocks (in Czech), Charles University, Prague.

Matolín, M., 1994: Radon risk determination of the geological basement (in Slovak). Manuscript, Uranpres, Spišská Nová Ves.

Murith, C. & Gurtner, A., 1990-93: Mesures in situ etirradiation externe, In: "Environmental Radioactivity and Radiation Exposure in Switzerland (Annual Reports 1990,1991, 1992, 1993, BAG Bericht, Bundesamt für Gesundheitswesen, Abteilung Strahlenschutz, Bern, Switzerland.

Polánski, A. & Smulikowski, K., 1971: Geochemistry (in Slovak). SNTL Bratislava