

NATURAL OCCURRING URANIUM IN SERBIA AND POSSIBLE ENVIRONMENTAL EFFECT

B. RADOSEVIC^a, J. KOVACEVIC^b, V. JOVIC^c

^a "Advanced Systems", 18 Vojislava Ilica Street, 11 000 Belgrade, Yugoslavia

^b "Geoinstitut", 12 Rovinjska Street, 11 000 Belgrade, Yugoslavia

^c Faculty of Mining and Geology, 7 Dusina Street, 11 000 Belgrade, Yugoslavia

E-mail: miki@opticom.co.yu

Abstract. Uranium investigations were carried out for decades, after the Second World War, in Serbia. They were terminated in the early 1990's for various reasons, mainly economic ones. These investigations yielded the "Kalna mines", which were in operation from 1996 to 1999, as well as several noneconomic deposits. This paper deals with the numerous collected data from environmental point of view. The idea of the work is to point out to the areas which could be of environmental or health concern due to natural radioactivity in building stone, high ground radon emanation, discharge of acid water from abandoned mines and adits, radioactive dumps and tailings, etc.

Keywords: uranium, radon, uranium deposit, environment, Serbia.

AIMS AND BACKGROUND

Uranium investigations in Serbia commenced in 1948 and lasted, with short intermissions, up to 1990, when they were finally terminated. It is not very likely that they will be resumed, as the law, which forbids construction of nuclear power plants, was legislated in 1989. The first period of investigations was accompanied with great secrecy as at that time it was treated as strategic mineral raw material, while in the second period, coinciding with the ending of the cold war, it was investigated as energetic raw material for nuclear power plants when the main investor was the government electric power company. During some 40 years of investigations numerous data were collected, by various prospecting methods, which can be well correlated as they were collected by the only one government company "Geoinstitut" from Belgrade, analysed and measured with the same laboratory and field instruments, by the same specialists and analysts. Sources of information are diverse, from aerogammaspectrometric data and ground checking of airborne anomalies to geochemical data from methods like hydrogeochemistry, panned concentrates, soil sampling and gas emanations.

Uranium is one of important elements to monitor in order to protect the environment from its deleterious influence. As an element it is a toxic heavy

* For correspondence.

metal, although reported intoxications are extremely rare. Its radioactive action is also highly harmful to living world, but high expositions are very rare in nature, confined only to artificial ambients like laboratories, nuclear power plants, etc. However, Rn^{222} , a product of the decay chain of naturally occurring U^{235} , is a pollution concern both in air and water, with established cancerogenic properties. Limited investigations in Serbia showed that this problem definitely exists^{1,2}.

This work is a presentation of all known uranium occurrences, in an increased quantity, on the territory of Serbia where possible environmental problems may occur due to natural contamination as well as due to higher radon emissions. Artificial occurrences like low grade ore dumps, exploration addit dumps and radioactive water discharge from abandoned mines and exploration addits are also presented in this work.

URANIFEROUS FORMATIONS IN SERBIA AND TYPES OF DEPOSITS

There are several identified uraniferous formations in Serbia which were investigated in detail. They are the following :

- **granitoid rocks** which are the host rocks of the largest number of occurrences and deposits. All of these deposits are of hydrothermal type. The age of the granites ranges from Palaeozoic to Late Tertiary. There are several granite complexes which are bearers of these mineralisations Janja with Mezdreja, Gabrovnica and Srnci Do deposits, Bukulja with Paun Stena and Cigankulja deposits, Cer, Kukavica, Jastrebac and Slatinska Reka. The most important deposits are in Gabrovnica and Mezdreja, the "Kalna mines", which were in production in the period of 1960-1966;

- **crystalline schists** are of diverse lithologic composition and various stage of metamorphism, usually older than Upper Carboniferous. They compose a large area of Serbia, but with more or less insignificant mineralisations. These mineralisations are mostly of infiltration type (Trepetljak, Klokočevac, Turija) but some are of hydrothermal origin (Nekudovo, Resavica);

- **volcanic rocks** of Pre-Cretaceous and of Tertiary age. The only deposit in Serbia, belonging to this formation is Muhovo. The deposit is of hydrothermal origin;

- **pre-Tertiary terrigene sandstones** represent the most significant bearers of mineralisations in Serbia. Two deposits, Dojkinci and Plavna, were discovered, while several other mineralisations were investigated - Lokve, Senokos, Porečka Reka. These deposits are of infiltration type;

- **tertiary sediments** are also frequent bearers of mineralisations in Serbia. The mineralisations are usually confined to small, isolated, basins composed of clastic sediments of Miocene to Pliocene age. They were discovered in Iverak and Vranje basins as well as a deposit in Belanovica basin.

It should be noted that the investigations were conducted under centrally planned government budget and that practically all of these deposits (Fig. 1) are uneconomic in view of market based economy. The cut-off grade adopted at that time by the government entities was 300 g/t. For this reason, apart from the environmental issues, it is highly improbable that investigations will be resumed in near future. The grade/tonnage of the deposits are given in Table 1.

Table 1. Grade and tonnage of deposits in Serbia

Deposit	Category	Tonnage (t)	Grade U_3O_8 (%)	U_3O_8 (t)
A. Janja granite				
Mezdreja	B	235 116	0.043	100
	C1	520 214	0.031	161
	B+C1	755 380	0.035	261
	C2	277 414	0.029	79
Gabrovnica	B	68 571	0.035	24
	C1	54 000	0.030	16
	B+C1	122 571	0.032	40
	C2	39 000	0.040	16
Srnci Do	C1	115 000	0.040	46
	C2	140 000	0.040	56
	D1	3 067 567	0.037	1135
B. Stara Planina Permian-Triassic				
Dojkinci	C2	1 800 000	0.064	1150
	D2	2 000 000	0.060	1200
C. Plavna Liassic				
Plavna	C2	93 333	0.075	70
	D1	160 000	0.050	80
D. Bukulja granite				
Cigankulja	C2	1 338 510	0.063	837
	D2	121 000	0.044	53
Paun Stena	C2	3 366 181	0.033	1110
E. Belanovica Tertiary basin				
Srednje Brdo	C2	1 231 680	0.061	756
F. Iverak Tertiary basin				
Ribarice	C1	711 953	0.029	207
	C2	691 175	0.022	154
	D1	2 400 000	0.025	600

HYDROGEOCHEMICAL DATA

The surface and ground waters of Serbia generally have somewhat higher content than in Russia and USA, for example. According to Durrance³ Russian rivers (100 investigated) contain 0.07 to 7.00 ppb, mean value 0.50 ppb, while USA

rivers have a mean content of 0.1 ppb. In uraniumiferous areas the contents range from 1 to 10 ppb in surface and 1 to 100 ppb in ground waters, while mine waters contain from 15 to 400 ppb. In Serbia the mean content is 1-2 ppb. Within Bukulja granites streams have mean content of 3 ppb while wells have mean value of 13 ppb. In Cer granites all waters contain about 0.7 ppb⁴. The thermal baths in Serbia used for radiotherapy contain up to 200 Bq/l of Rn and up to 0.481 Bq/l of Ra⁵.

Hydrogeochemistry was applied in most of the investigated areas. Waters of the Tertiary basins are in places with very high Rn content. In the Iverak Tertiary basin there is a spring (Majur-Slatina) with extraordinarily high radon content of 350 – 3600 Bq/l (measured over a period of 11 years), Ra from 0.711 to 0.739 Bq/l and extremely low U (Ref. 6). The Vranje area has a high mean content (500 samples) of Rn 7.025 Bq/l, ranging from 0.850 to 70.000 Bq/l, while the Ra and U values are within background values of the Tertiary basins ranging from 0.010 to 0.190 Bq/l (mean 0.037 Bq/l) and 0.20 to 210.00 µg/l (mean 1.71 µg/l)⁷. In the Vrsacki Bregovi area (330 samples) Ra ranges from 0.05 to 1.17 Bq/l, mean value 0.17 Bq/l, Rn ranges from 1.77 to 122.93 Bq/l (mean value 12.70 Bq/l) and U ranges from 0.2 to 120 µg/l (mean value 1.23 µg/l)⁸. The high values of U, in places, could be a consequence of the use of Morocco imported radioactive phosphate fertilizers that were frequently applied. In the Cer-Iverak area 60 samples were taken for analyses⁴. The mean value of U was 1.2 µg/l ranging from 0.1 to 22 µg/l. The one anomalously high value was found in Ravni Potok which flows over U mineralisation (300 g/t). An extremely high anomaly was encountered during drilling for U in Bukulja granite, Rn 11.000 Bq/l and U 200 g/t (Ref. 9). In the Barbeš Tertiary basin 127 samples were taken from streams, wells and springs with 0.3 to 214 ppm U (mean 2.7 ppm), 0.004 to 0.307 Bq/l Ra (mean 0.051 Bq/l) and 3.33 to 259.00 Bq/l Rn (mean 20.42 Bq/l) (Ref. 10). Subsequent investigations within Barbes basin yielded slightly higher results of Rn, up to 377.4 Bq/l, much higher Ra values ranging from 0.109 to 0.794 Bq/l (mean 0.300 Bq/l) and slightly lower U values¹¹. The highest Rn value of 5020 Bq/l was measured in a spring

Table 2. Water samples from Belanovica Tertiary basin¹³

Sample source	Ra (Bq/l)	Rn (Bq/l)	U (µg/l)
Spring	0.220	30.410	0.6
Tap	0.246	7.515	2.5
Spring	0.135	20.551	0.5
Tap	0.054	4.242	0.3
Spring	0.273	31.834	0.3
Spring	0.054	1.985	0.3
Spring	0.083	32.794	0.4
Lake Garaši	0.082	8.158	-

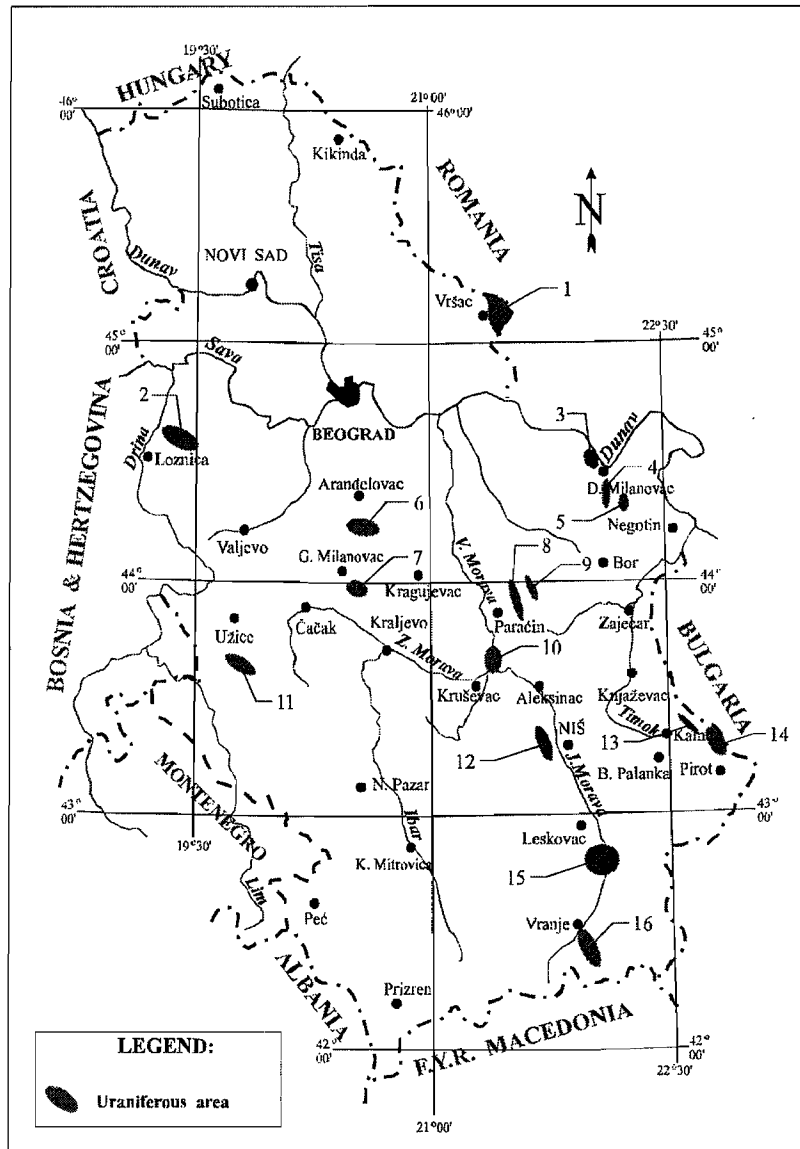


Fig. 1. Uranium-bearing areas of Serbia and type of mineralisation: 1 – Vrsacki Bregovi, crystalline schists hydrothermal; 2 – Cer-Iverak, granite (Cer) hydrothermal and Tertiary sediments (Iverak) infiltration; 3 – Lepenski Vir, Pre-Tertiary terrigene sandstones infiltration; 4 – Porecka Reka, pre-Tertiary terrigene sandstones infiltration; 5 – Plavna, pre-Tertiary terrigene sandstones-infiltration; 6 – Bukulja-Belanovica, granite (Bukulja) hydrothermal, Tertiary sediments (Belanovica) infiltration; 7 – Takovo, volcanic rocks hydrothermal; 8 – Resavica, crystalline schists-hydrothermal and infiltration; 9 – Nekudovo, crystalline schists hydrothermal and infiltration; 10 – Stalac, crystalline schists hydrothermal; 11 – Muhovo, volcanic rocks hydrothermal; 12 – Jastrebac, granite and crystalline schists hydrothermal; 13 – Janja, granite hydrothermal; 14 – Dojkinci pre-Tertiary terrigene sandstones infiltration; 15 – Slatinska reka, granite hydrothermal; 16 – Vranje, Tertiary sediments infiltration

near Stalas in Pre-Cambrian crystalline schists¹². However, out of over 100 samples only 3 samples had higher values of U, Ra and Rn. The data for surface waters from Belanovica Tertiary basin are presented in Table 2. Waters from the drill holes in Belanovica basin yielded up to 0.410 Bq/l of Ra, 97.375 Bq/l of Rn and 7.000 µg/l of U.

EMANATION DATA

Another frequent prospecting method that was applied in the investigation was emanation. Most of the projects incorporated this method. In some areas high emanation values in relation to normal Rn content of about 20 Bq/l were obtained. Data for some areas in Serbia are given in Table 3.

Table 3. Emanation values for some areas in Serbia

Area	Number of points	Max. value (Eman)*	Mean value (Eman)	Standard deviation	Coefficient of variation
Stara Planina ¹⁴	177	20	-	-	-
Stara Planina ¹⁵	5084	37	7.8	4.2	-
Plavna ¹⁶	250	44	10.4	6.1	58.3
Nekudovo-Resavica ¹⁷	108	30	8.5	-	-
Toplica basin ¹⁸	360	13	7.6	-	-

* 1 Eman equals to 3.7 Bq/l.

STREAM SEDIMENT DATA

In the Vranjska Banja area 2230 stream sediments were collected from the streams within this Tertiary basin. The values range from 0.40 to 8.00 g/t, mean value 1.67 g/t (Ref. 7). In the Cer-Iverak area 60 samples were taken for analysis⁴. The content in these samples ranges from 1.5 to 3.0 g/t with an exception of one sample where 55 g/t was established. This sample was taken from Ravni Potok which was mentioned in hydrogeochemistry section. From the streams of Stara Planina, flowing over Janja granites, over 4000 samples were collected. 150 samples were compared on content in stream sediments and granite rock samples, where mean value of stream sediments was 10.6 g/t and rock samples 6.4 g/t (Ref. 19). In the Leskovac area 276 samples were taken with 4 to 35 g/t, mean 8.7 g/t (Ref. 10).

PANNED CONCENTRATE DATA

Usually panned concentrates were collected to mineralogically determined presence of particular interesting minerals. In some cases they were analysed chemically for U and Th. In the Bukulja area 61 samples had mean content of 160.9 ppm U and 933.5 ppm Th (Ref. 20).

SOIL DATA

Soil sampling was rarely applied in prospecting in Serbia. In the Vrsacki Begovi area 937 samples were collected. The values range from 0.40 to 2.50 ppm, mean value 0.70 ppm (Ref. 8).

ROCK DATA

The rocks in Serbia generally have higher content of U than Clarke for the same rocks. Numerous data were collected during years of U exploration and only selected data will be presented. The Clarke for some common rocks is given in Table 4, while in Tables 5, 6 and 7 data for some rocks in Serbia are given.

Table 4. Clarke for U and Th of some common rocks

Rock	U (ppm)	Th (ppm)
Meteorites ²¹	0.009	0.038
Earth's crust ²²	1.500	7.300
Basic rocks ²²	1.000	4.000
Acidic rocks ²²	3.000	17.000
Sandstones ²²	0.450	1.700
Schists, shales ²²	3.700	12.000
Limestones ²²	2.200	1.700
Sea water ²¹	0.003	0.00001

Table 5. Average U and Th content of some igneous rocks of Serbia

Locality rock	U (ppm)	Th (ppm)
1	2	3
Timok-Pek andesite ⁴	1.3	4.6
Gnjilane trachytoid ⁴	11.7	49.0
Cer granite ⁴	4.5	13.0
Bukulja granite ⁴	4.3	15.0
Janja granite ⁴	6.4	29.0
Bujanovac granite ⁴	1.9	4.5
Golija granite ²³	6.6	23.8
Golija granodiorite-porphyrityte ²³	12.9	21.5
Borac quartzlatite ²⁴	7.8	29.7
Borac andesite ²⁴	6.9	26.3
Borac pyroclastics ²⁴	10.5	30.1
Brajkovac granodiorite ²⁵	4.7	9.6
Kukavica granite ²⁶	4.5	11.9
Besna Kobilja granodiorite ²⁶	6.7	18.0
Cer granodiorite ²⁷	4.7	14.0
Boranja granodiorite ²⁸	5.9	19.9
Zeljina granodiorite ²⁹	4.5	16.3

to be continued

Continuation of Table 5

1	2	3
Kopaonik porphyroid granodiorite ³⁰	9.1	29.6
Rudnik dacite ³¹	6.7	21.6
Kotlenik andesite ³²	2.4	9.2
Kotlenik pyroclastics ³²	4.3	12.5
Jastrebac granodiorite ³³	4.9	12.4
Surdulica granodiorite ³⁴	6.8	17.8
Surdulica quartzlatite ³⁵	7.2	22.3
Surdulica dacite ³⁵	5.4	17.8
Slatinska Reka granite ³⁶	5.5	13.1
Lece andesite ³⁷	5.5	13.1

Table 6. Average U and Th content in some sedimentary rocks (surface and drill holes data) of Serbia

Locality rock	U (ppm)	Th (ppm)
Stara Planina gray-reddish fine-grained sandstone ³⁸	18.0	17.0
Stara Planina gray middle-grained sandstone ³⁸	45.0	9.0
Stara Planina red siltstone ³⁸	5.0	16.0
Stara Planina gray sandstone with siltstone fragments ³⁸	64.0	13.0
Stara Planina dark gray silty and fine-grained sandstone ³⁸	47.0	18.0
Stara Planina altered and bleached sandstone ³⁸	53.0	11.0
Stara Planina red silty and fine-grained sandstone ³⁸	9.0	16.0
Stara Planina red middle-grained sandstone – base of “varicolored sandstone series” ³⁸	3.0	11.0
Plavna tuff ³⁹	4.0	9.1
Plavna tuffite ³⁹	5.2	11.2
Plavna sandstone ³⁹	4.2	8.1
Plavna tuff-breccia ³⁹	5.1	9.1
Plavna red ferruginous limestone ³⁹	1.2	8.2
Vladicin Han tuff ⁴⁰	6.7	21.4
Pocuta bauxite ⁴¹	4.7	27.7
Mackat bauxite ⁴²	4.9	25.5
Mackat clayey bauxite ⁴²	2.7	17.0

Table 7. Average U and Th content in some metamorphic rocks of Serbia

Locality rock	U (ppm)	Th (ppm)
Kukavica gneiss ²⁶	2.8	11.9
Kukavica two mica schist ⁴³	2.0	4.8

OTHER DATA

In 1992 geocological concern was expressed by the municipal authorities in Knjazevac, a town nearest to the former mines. By their request preliminary radioactive survey of the area was performed by “Geoinstitut”, the same company that investigated the area for several years back⁴⁴. After five years some of

the methods were again applied to determine whether the situation has changed⁴⁵. Unfortunately, due to the limited funds only a small number of samples were collected so that firm conclusions could not be drawn. However, some interesting results were obtained.

Samples of water that is discharging from the abandoned mines have shown significant increasing of U content after five years (Table 8). It is difficult to explain this increase on the basis of only two water samples in the period of five years, but it is possible that there is an inflow of mildly acidic waters which are more efficient in leaching.

Hydrogeochemical samples taken several kilometers downstream did not exceed content of 0.005 mg/l. The same situation is with the wells from the village Balta Berilovac, a few kilometers downstream from Mezdreja mine. This is to be expected due to high migrativity of UO_2^{2+} ion which would migrate far downstream and could be precipitated only in porous sedimentary material containing organic matter. For that reason humus near the banks of the streams and stream sediments were analysed (Table 9), as well as water from the village wells situated in alluvial deposits of the same streams (Table 10). The normal background of U in humus in the area is 1 ppm. It is hard to determine whether the somewhat higher values that were found in the samples are due to the contamination or the due to natural weathering of host rocks of the area (Janja granites and Inovo metasediment series) which have an increased natural U background of 4 ppm. However, the water samples express low values which leading us to the conclusion that there is only limited contamination by U, from the abandoned mines, in the area.

Table 8. Hydrochemical analyses of mine discharge water⁴⁵

Year	U (μ g/l)	Ra (Bq/l)	Fe (mg/l)	Sr (mg/l)	Pb (mg/l)	Zn (mg/l)	Cu (mg/l)	As (mg/l)	Minera- lisation	pH
Mezdreja mine										
1992	93.8	0.07	0.37	4.40	<0.005	0.04	<0.01	0.014	470	7.4
1997	420.0	0.32	1.08	12.38	<0.010	-	<0.01	-	860	7.5
Gabrovnica mine										
1992	27.2	4.42	0.13	0.42	<0.020	N/A	<0.01	<0.005	622	6.8
1997	50.0	1.50	0.56	0.48	<0.010	N/A	<0.01	-	720	6.9

Hand scintillometric survey showed that only the area in the perimeter of several tens metres from the mines and dumps has high scintillometric count, gradually decreasing getting further away from these objects.

The Rn emanation values are high within the low grade ore dump, and the vicinity of the former processing plants where some grinded material was left after the closure of the mine.

Table 9. Radiometric analyses of stream sediment and humus samples downstream from the mines⁴⁵

Type of sample	U (ppm)	Th (ppm)	K (%)	Th/U
Stream sediment	3.79	9.66	1.88	2.54671
Stream sediment	2.30	10.24	1.91	4.46154
Stream sediment	4.01	9.65	1.70	2.40522
Stream sediment	4.33	11.12	1.87	2.57055
Humus	4.88	12.16	1.71	2.49065
Stream sediment	3.03	5.49	2.04	1.81137
Stream sediment	3.03	6.41	1.60	2.11429
Humus	3.13	8.42	1.58	2.69331
Stream sediment	2.94	7.35	1.62	2.49475
Stream sediment	5.01	13.33	1.77	2.66222
Stream sediment	4.28	9.01	2.12	2.10500
Stream sediment	2.28	7.05	1.70	3.09698
Stream sediment	2.05	6.28	2.06	3.05691
Humus	3.06	10.20	1.59	3.33123
Stream sediment	2.21	8.17	1.51	3.68992
Humus	3.37	12.25	1.37	3.63495
Stream sediment	3.60	12.03	1.80	3.34631
Stream sediment	1.38	3.38	0.68	2.45364
Stream sediment	3.23	5.28	1.12	1.63214

Table 10. Analyses of water samples from the alluvial deposits downstream of the uranium mines⁴⁵

Locality	U ($\mu\text{g/l}$)	Ra (Bq/l)	Fe (mg/l)	Sr (mg/l)	Pb (mg/l)	Cu (mg/l)	Minera- lisation	pH
Gornja Kamenica well	2.00	0.60	2.15	0.15	<0.01	<0.01	1020	7.3
Donja Kamenica tap	1.00	0.10	0.06	0.21	<0.01	<0.01	570	7.8
Štrbac well	1.00	<0.05	0.22	<0.01	<0.01	<0.01	900	7.3
Baranica tap	1.00	0.20	0.97	<0.01	<0.01	<0.01	360	7.6

CONCLUSIONS

Numerous data, collected during uranium exploration, clearly indicate that there are areas where there is a possibility that natural radionuclides can endanger human/animal health. This can specially be stressed in areas where there are settlements over, or nearby mineralisations and larger occurrences. This can also be a problem in areas where there is tradition to use building stone for building houses⁴⁶. Hazardous point areas are identified in places where there is no municipal water, and the springs or wells used for water supply contain radionuclides in higher content than the drinking water standards. This paper represents a synthesis of all known data, pointing to areas that should be investigated in detail to check the actual natural radionuclides influence onto the living world.

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