

## GEOECOLOGICAL RADIOACTIVE RISKS IN THE AREA OF BORANJA GRANODIORITE MASSIF

**V. Gordanic, D. Jovanovic, A. Ciric**

*GEOINSTITUTE, Rovinjska 12, 11 000 Belgrade, Serbia and Montenegro*

**ABSTRACT.** Boranja granodiorite massif is placed in western Serbia where it spreads at about 80 km<sup>2</sup>. Geoecological state in this area, caused by Boranja granodiorite, is based on results of petrological, geochemical and structural investigations.

The main part of the pluton is intruded in the dome of anticline made of Permo-Carboniferous and Mesozoic formations.

During the intrusions ring structures and fault zones striking NW-SE were formed. They, as well as fractures in the denudated parts of massif, represent migration paths for radioactive elements from magmatic body.

In order to define radioactive risks we have determined the amounts of U, Th and K from rock samples, content of total and soluble U from the stream sediments and content of U, microelements, anion-cation composition, pH, Eh and Ep of the waters.

Geoecological significance of obtained results is especially important for space planning in waterpower engineering and agriculture as well as in adequate protection of inhabitants and living world in general.

## ГЕОЕКОЛОЖКИ РАДИОАКТИВНИ РИСКОВЕ В РАЙОНА НА ГРАНОДИОРИТЕН МАСИВ БОРАНЯ

**В. Горданич, Д. Йованович, А. Сивич**

*Геоложки институт, ул. "Ровинарска" 12, 11 000 Белград, Сърбия и Черна гора*

**РЕЗЮМЕ.** Гранодиоритен масив Бораня се намира в западна Сърбия, където заема площ от около 80 km<sup>2</sup>. Геоекологичното състояние на района, дължащо се на гранодиоритен масив Бораня е проучено с помощта на петрологични, геохимични и структурни изследвания.

Основната част от плутона е внедрена в купола на антиклинала, изградена от карбон-пермски и мезозойски формации.

По време на внедряването са се оформили пръстеновидни структури и разломни зони, които имат простирание СЗ-ЮИ. Разломните зони, както и пукнатините в денудираните части на масива представляват миграционни пътища за радиоактивните елементи от магменото тяло.

За определяне на радиоактивния риск с определени количествата на U, Th и K в скални проби, съдържанието на общия и разтворения във седиментите U и съдържанието на U, микроелементи, съотношения аниони – катиони, pH, Eh и Ep на водите. Геоекологичното значение на получените резултати е изключително важно за пространственото проектиране на хидроенергийните съоръжения и обекти на селското стопанство, както и за съответната защита на населението и живите организми като цяло.

### Introduction

Boranja granodiorite massif is placed in western Serbia where it spreads at about 80 km<sup>2</sup>. Geoecological state of this massif was defined through the influence of natural radioactivity for the purposes of space planning of the urban places and waterpower engineering.

Definition of the level of radioactive safety in the ambience of this intrusive is based on the results of the content of natural radionuclides in different geochemical surroundings.

Geological mapping and sampling of the stones, stream sediments and waters was based on petro-structural characteristics of the terrain. Considering that the research took place in different mediums, the evaluation of the obtained results and definition of radiation safety was performed according to recommendations of the World Health Organization (WHO), International Committee for Radiation Protection (ICRP) and other international institutions.

### Main geologic features of investigated area

Investigated area included granitoid massif of Boranja and contact belt around it. It is bordered by the river Drina from the

south and west, river Radalj on the north and Mackov kamen on the east. Granodiorite is about 7 km wide and 12 km long. The highest parts of terrain are built of magmatic rocks, except the peak Mackov Kamen (924 m), which is built of shists and quartz sandstones. High tops are also Turski grobovi (783 m) and Alina glava (696 m). Rest of the terrain is characterized by the smooth relief which inclines gradually to the Drina valley.

Hydrographic network of this area is well developed and most of the rivers flow to the Drina.

Investigated area is characterized by complex geological composition. It is built of sedimentary rocks Paleozoic (Devonian-Permian); Mesozoic (Triassic-Jurassic) and Cenozoic (Miocene-Holocene) in age. Metamorphic rocks are Paleozoic and Tertiary in age, while volcanic and igneous rocks are from Tertiary.

On the eastern border of the massif is placed Drina-Ivanjica Paleozoic complex, while on the western side is found Diabase-Chert formation Jurassic in age. On the north and partly on the southeast (but in lesser amount) occur dacito-andesites. Their thickness is up to 100 m, and they are followed by pyroclastic material. They are porphyritic rocks with or without quartz, and with andesine and colored minerals - biotite, amphibole and pyroxene as phenocrysts. In these parts

we can also find quartz latites. They are Miocene-Pliocene rocks built of sanidine, quartz, andesine, biotite and sometimes amphibole.

The age of Boranja granodiorite is determined as Oligocene based on K/Ar analyses on hornblende and biotite.

Of the main interest for us were granitoid rocks and dyke rocks in them. Surface of about 80 km<sup>2</sup> occupied by granitoid was covered by detailed prospection, observation and sampling.

## Materials and methods

Activity of the natural radionuclides was determined on the samples from granodiorite massif, stream sediments and waters.

Massif itself consists mainly of amphibole-biotite granodiorites, with minor amounts of biotite granodiorites and monzonites. They are built of quartz, plagioclase, K-Feldspar, amphibole and biotite as main constituents with accessories apatite, titanite, zircon, orthite and metallic minerals.

Dyke rocks that follow intrusion are granodiorite porphyries, aplites, pegmatites and lamprophyres.

In granodiorite porphyries as phenocrysts occur quartz, orthoclase, plagioclase, biotites and amphiboles. Chemically, these rocks are very close to the main granodiorite intrusion.

Aplites and pegmatites are similar but differ in the size of the grains. They are built of quartz, K-feldspar, biotite, muscovite and accessories titanite and tourmaline.

Lamprophyres are very rare. According to their mineral composition, kersantite and minette are singled out.

Samples of the stream sediments and waters are gathered from the surface flows in accordance with hydrographic features of terrain.

Radiometric examinations were carried out on the samples previously mechanically crushed and powdered to 80 to 100 meshes and homogenized. Samples of about 500 g such prepared rocks were measured on gamma spectrometer with NaI(Tl) 4x4 in detector. For spectrum calibration and calculation of radioactive elements concentration, we have used "New Brunswick Lab." (USAEC): NBL No. 103 (0.05% U) as well as NBL No. 107 (0.10% Th) standards of uranium and thorium ores.

Determination of uranium in stream sediments and waters was performed by laser fluorimeter (LA UA-3) with fluran as characteristic reagent.

Chemical analyses of metal were carried out by following methods:

- Atomic Absorption Spectrophotometry (AAS)
- Atomic Emission Spectrophotometry (AES)
- Inductive Coupled Plasma- Atomic Emission Spectrometry (ICP-AES).

## Results and Discussion

Definition of geoeological status of Boranja granitoid massif refers to determination of zero-level of natural radioactivity in rocks, stream sediments and waters.

According to the results of radiometric analyses of rocks, amounts of U, Th and K vary: for uranium from 4.6 ppm to 8.65 ppm,  $x=6.3$  ppm; for thorium from 15.7 to 20.7 ppm,  $x=18.2$  ppm; for potassium from 2.3% to 4.1%,  $x=2.8$  %. Relation Th/U

ranges from 1.97 to 4.36 and indicates thorium character of investigated area.

According to mineral and chemical composition, the most of the examined rock samples correspond to granodiorites. Main oxide values, expressed in weight percents, vary within the ranges as follow: SiO<sub>2</sub> = 61.84-71.60%; Al<sub>2</sub>O<sub>3</sub> = 13.83 -16.76%; Fe<sub>2</sub>O<sub>3</sub> = 0.72-4.13 %; FeO = 0.62-2.3 1%; MnO = 0.05-0.10%; MgO = 0.71-2.50 %; CaO= 1.99-4.67%; Na<sub>2</sub>O= 2.64-3.65%; K<sub>2</sub>O = 2.14-4.05% and P<sub>2</sub>O<sub>5</sub> = 0.05-0.18%.

Trace elements analyses showed that, compared to average values for acid rocks, some of them have higher amounts such as: Ba (450-10000 ppm); Mo (7-25 ppm, rarely 60 ppm); Pb (10-70 ppm) and Sr (400-3000 ppm); while the content of others (Mn, Ga, V, Cu, Y, Yb, Zr, Ni, Co, Sc, Cr and La) correspond to the average values.

Genetic and geotectonic classification of Boranja granitoid rocks was based on their mineral and chemical composition. According to this, they are mostly I-type granitoides (Chappell&White, 1974; Hine *et al.*, 1979) and they have calc-alkaline character. That was confirmed in the papers of Steiger *et al.*, 1989; Karamata *et al.*, 1990; Knezevic *et al.*, 1994.

According to mentioned authors, at the beginning of Oligocene partial melts from the upper mantle were moved towards the surface and mixed with the material from the lower parts of the continental crust. This lifting occurred through the main, big fractures striking NNW-SSE (striking of Vardar Zone). During this process, chemical composition of the melt has been continuously changing from quartzmonzonite to granodiorite.

Boranja granitoid massif is characterized, based on the morpho-structural analyses, by clear differentiation of massif itself, unequal erosion, local reefs, sharp linear discontinuity in NW-SE part of the massif, which is in the main part of the terrain separated from the Paleozoic shists, and numerous fractures and ruptures.

The big part of the massif is gneissified, on some places its thickness is even 5 m, especially in the parts with small dip.

In stream sediments, which was made by fluvial erosion and denudation of this massif, determined contents of uranium varies within range from 0.2 to 12 ppm,  $x = 1.2$  ppm. According to Atlas BRGM, the average content of U in stream sediments is 0.5 ppm. It implicate that it would be necessary to analyze sand quarries by the rivers especially if the material possibly origins from the areas of the increased radioactivity.

In the massif itself, some aquifers connected to the fault structures and fractures are formed in the different depths. They are the cause of occurrences of thermal waters in Radeljska Banja (Spa) which is used for drinking and bathing.

Uranium content in underground and surface waters varies within range from 0.01 to 2.0 ppb,  $x = 0.8$  ppb with pH values 6.4 to 8.2. Waters are slightly mineralized, neutral to slightly alkaline. According to anion and cation content, they mainly correspond to hydrocarbon-chloride-sulfatic, hydrocarbon-sulfatic-chloride, sodic-potassic-calcic-magnesian to sodic-magnesian-calcic type. This kind of waters with slightly alkaline reaction (pH=7.5-8) is very suitable for dissolution of carbonates, sulfates, vanadates and uranium oxides. Regarding the degree of cationic influence to uranium leaching from surrounding rocks, on the first place is Na followed by Ca and Mg.

Utilization of Boranja granodiorite as building material and in architecture was determined, among other things, by the index

of total activity (I) which is in the about 30% of the examined samples > 1 or at the border (Table 1).

For maintaining the level of the radiation safety, most of the European countries have brought regulations about permitted amounts of natural radionuclides in building materials, as well as recommendations for the level of radon concentrations especially in living and working spaces. According to this recommendations, total index of gamma radiation of natural radioactive elements uranium, thorium and potassium in building material should not exceed  $I = 1$  (0.7 mSv/year) and interventional level of radon in living spaces should not be higher than 400 Bq/m<sup>3</sup> in old, and 200 Bq/m<sup>3</sup> in new buildings.

Table 1.

Content of radionuclides in granodiorite samples of Boranja

sample	<sup>226</sup> Ra Bq/kg	<sup>232</sup> Th Bq/kg	<sup>40</sup> K Bq/kg	γ-Index
1	98.89	77.05	782.18	>1
2	75.89	78.99	1226.12	>1
3	81.51	77.58	824.46	<1
4	64.26	74.83	854.66	<1
5	76.14	66.25	794.26	<1
6	88.77	70.86	818.42	<1
7	62.76	83.68	1078.14	<1
8	68.76	77.49	860.70	<1
9	73.26	73.69	748.96	<1
10	91.64	79.23	854.66	>1
11	108.14	68.96	839.56	>1
12	65.51	76.56	821.44	<1
13	71.89	63.58	939.22	<1
14	57.76	65.28	779.16	<1
15	35.13	34.22	513.40	<1
16	38.26	53.91	918.08	<1
17	101.77	71.06	776.14	1
18	82.52	83.40	890.90	<1
19	89.77	70.62	673.46	<1
20	78.14	72.36	742.92	<1

Geoecological risk refers to the exposure of the population to the ionizing radiation according to recommendations of World Health Organization, United Nations Commission for Atomic Energy Research (UNCAER) and other international and national institutions.

For example, we mention the place G. Stubla in South Serbia, where the measured concentrations of radon in the houses were higher than 3000 Bq/m<sup>3</sup> (on some places even up to 9000 Bq/m<sup>3</sup>, measured by detectors CR-39 type). Index of total gamma activity of trahites used for building of this houses is >1.

## Conclusion

According to the presented results we can make following conclusions:

- It is very important to determine geoecological status, zero-level of natural radioactivity of the area in which, eventually, is planned building of some objects.
- It is necessary to carry out existing regulations more severe and establish permanent control of building material, especially decorative stones, domestic as well as imported.
- It is very important to single out the zones of radiation-radioactive risks including contents of natural radionuclides in different environments.
- In the areas of anomaly concentrations of radioactive elements in the stones, earth, air and water, in the working and living places, it is necessary to carry out adequate health protection of inhabitants.

## References

- Chappell B. W. & White A. J. R., 1974: Two contrasting granitic types, *Pacif. Geol.*, 8, Tokyo, 173-174.
- Fisenne I. M.: *Long-lived (natural) Radionuclides in the Environment*. In Final Report from V<sup>th</sup> Inter. Symp. On Natural Radiat. Environm. EUR 14. 411, CEC, p 187-255, Luxemburg, 1993.
- Gordanic V., Jovanovic D., 1998: *Natural radioactivity of building stone from selected quarries in Serbia*. Symposium of measurement and measurement equipment. Proceedings, book II, p 771-777, Belgrade 1998. (in Serbian)
- Gordanic V., Jovanovic D., Zunic Z., 1998: *Radionuclides in building stone with a reference to Indoor Radon Concentration Level*. XIII congress of geologists of Yugoslavia, book IV, mineral raw materials, p 327-333, Herceg Novi, 1998. (in Serbian)
- Hine R., Williams I. S., Chappell B. W. & White A. J. R., 1979: *Contrast between I- and S-type granitoids of the Kosciuszko batholithe*, *Jour. Geol. Soc. Australia*, 25, 219-234.
- Karamata S., Steiger R., Djordjevic P. & Knezevic V., 1990: *New data on the origin of granitic rocks from Western Serbia*. Ext. du Bull. De L'Acad. Serb. Des Sci. et Arts, T. CII, Class. Des SCI. math. et natur., Sci. natur. No 32, 1-9. Beograd.
- Knezevic V., Karamata S. & Cvetkovic V., 1994: *Tertiary granitic rocks along the southern margin of the Pannonian Basin*, *Acta Mineralogica-Petrographica* XXXV, 71-80. Szeged.
- National Research Council Committee on The Biological Effects of Ionizing Radiation: *The Health Effects of Radon and other Internally Deposited Alpha Emitters*, book IV, Washington, DC: National Academy Press, 1988.
- Source and Biological Effects of Ionizing Radiation, UNCAER Publ., New York, 1988.
- Steiger R., Knezevic V. & Karamata S., 1989: *Origin of some granitic rocks from the southern margin of the Pannonian Basin in Western Serbia, Yugoslavia*. Terra Abstracts, vol. 1. 53, Strasbourg.