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# GEOCHEMICAL INDICATORS IN IDENTIFICATION OF GEOPATHOGENIC ZONES OF NATURAL RADIOACTIVITY

# GEOHEMIJSKI INDIKATORI U IDENTIFIKACIJI GEOPATOGENIH ZONA PRIRODNE RADIOAKTIVNOSTI

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*Ključne reči*

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*Abstract*

Identification of geopathogenic zones is based upon the data obtained from regional investigations of nuclear materials at various prospecting stages of geochemical areas and they are related to concentration of U, Th, K in soils and rocks as well as of U, Ra, Rn in water and Rn in air. Geochemical maps based on geostructural features of the terrain enable evaluation of external (ERB) and internal (IRB) human radiation burden. Presented results necessary for radiogeochemical endemic investigation (RGHE) of possible geopathological zones were obtained from Carpathian-Balkan counties (Serbia and Macedonia).

## *INTRODUCTION*

Exploration of nuclear raw materials in Serbia (Yugoslavia) have started in 1948 and lasted until 1990 with variable intensity and in several stages. After that, it continued with less intensity and with a significantly lower environmental and medical importance. Various methods were used during investigations as: remote sensing (application –processing of satellite and photo geological images), airborne gamma spectrometry, geochemical, geophysical, petrological, mineralogical, radiometric, chemical and other methods. Hundreds of thousands data were analyzed and divided into several groups depending on the importance for specific research area of medicine, agriculture, forestry, waterpower engineering, spatial planning and environmental protection. Results of geochemical prospecting of different milieus of geosphere were partly used for identification of geopathogenic zones [1,2] We continued to analyze the obtained data through several scientific and research projects from 2008 and here we present only a small part of our research as an idea which will contribute to further geomedical investigations.

## *MATERIALS AND METHODS*

### *Geographical focus*

Several areas were selected for investigation: Stara Planina massif (Janja granitoide) because the uranium mines Gabrovnica, Mezdreja and Srneći Do were found in vicinity of settlements; then Bukulja (central part of Serbia – Šumadija) and Stublovača (South Serbia) massifs, where nuclear raw materials have been investigated for many years and stone from quarries used for interior decoration.

For identification of geopathogenic zones it is significant to determine the intervention level of radon concentration not only in the air indoors, but also in waters, and to define the sources of natural radioactivity, which are of special significance for evaluation of external (ERB) and internal (IRB) radiation burden on human population. Radon (Rn) gas was used as immediate indicator of uranium deposits, ore occurrences and mineralizations. It is highly migrative in dependence of geological-structural features of the terrain, level of ground waters and surface waters, atmospheric pressure and other physical parameters[3].

Regionally, following successive phases of investigation, selected locations are in accordance with metallogenetic potential of the area for discovery of uranium deposits and ore occurrences, therefore they represent potential spaces of geopathogenic zones. Taking in the natural radionuclides by ingestion and inhalation was noted in the area of geopathogenic zones, but it is also necessary to emphasize that housings were mainly built of the materials from these areas, which cause the increase of external and internal radiation burden, as it is for example in the area of Bukulja and Stublovača.

### *Sampling and Instrumentation*

Contents of natural radionuclides were determined in the samples of water and rocks, and indoor radon concentrations were measured in housings. Samples of geological materials were treated by mixture of HNO3, HclO4 and HF acids (1:2:4). Intensity of complex fluorescence (formed by adding of Fl ) was measured from the aliquot taken from the filter. Analyzing was carried out by the method of standard adding. Radium was measured after 21 day. In the samples of water from the area of Bukulja massif, contents of uranium were determined by laser fluorimeter UA-3. Further, emanometry (emannometer RDU-200) was used for determination of Ra and Rn concentrations.

Rock samples were taken from Bukulja granitoide massif, from active and non-active quarries, and from the vicinity of radioactive anomaly Stublovača, after previous radiometric profiling (by scintillation detector GR-110). Rock samples with low uranium (< 20 ppm) and thorium (< 50 ppm) contents were previously milled (to 80-100 mesh) and homogenized. Samples of 300 g and 500 g were packed in special containers and measured by gamma spectrometer in radiometric laboratory. Measurements were carried out by apparatus that consist of scintillation detector "Bicron" with NaI(Tl) crystal 4"x4" and multichannel analyzer (MCA; 4096 channels) type EG&G "ORTEC"-7500. Analyses were based on measurements of high-energy radiations (up to 3 MeV) in certain parts of the spectrum. As the result, values in ppm (g/t) for U (total) and $^{232}$Th and in wt.% for total K, were obtained. Calibration constants obtained by measurements of gamma spectrum of standards were used for calculation of unknown concentrations. For spectrum calibrations and calculation of concentration of natural radionuclides, we used uranium and thorium ore standards from "New Brunswick" laboratory (USAEC-United States Atomic Energy Commission). Potassium chloride (p.a.) was also used as potassium standard.

In the samples with increased U, Th and K contents, analyses were repeated within low-energy part of the spectrum (up to 500 keV). Homogenized samples (100 g; ground to 100-150 mesh) packed into smaller containers were used for these analyses. Scintillation detector NaI(Tl) 2"x1" was used in the case of increased U, Th and K contents .

Radon concentrations ($^{222}$Rn) were measured indoors (in vicinity of Kalna and Gornja Stubla) by integral method in diffused chambers with solid trace detector CR-39 on the base of polycarbonate. Detectors were placed on the bottom of diffuse chamber in order to eliminate dust and radon daughter products. Exposure time of diffuse chambers was about 90 days. Detectors were developed chemically by using 6 M solution of KOH on the temperature of 70ºC, within 5 h, and then washed out in running water for 30 minutes. Readings of the á-particle's traces were carried out by optical microscopy[4].

## RESULTS AND DISCUSSION

Athmogeochemical and Geopathological map of Rn indoor concentration were made on the basis of the results of geochemical and radiological investigations of the Kalna area. Geological-structural characteristics of the terrain and uranium deposits Gabrovica, Mezdreja and Srneæi Do were presented in the basis of the map by compilation of existing data. Source material for formation of the uranium deposits and ore occurrences were Janja granites, metasediments of Inovo series and Permo-Triassic terrigenous sediments. All deposits were placed around the place Kalna, and their common characteristic was that they are connected with fault structures. Ore mineralizations were in the shape of lenses, veins and columns[4].

Smaller settlements Gabrovnica, Donje Polje, Stara Kalna, Inovo, Vrtovac, Balta Berilovac, Mezdreja, Janja and Ravno Buèje were located around uranium deposits. Indoors radon concentrations were measured at the housings of these settlements in the periods: March-July: I quarter, July-October: II quarter, October-February: III quarter and February-June (next year): IV quarter. Measurements were carried out in 78 houses with 104 pieces of diffuse chambers with CR-39 placed in the living rooms and bedrooms.

Radon concentrations varied during the seasons. Results of radon concentration are presented in Table 1.

| Quarter | Maximal value [Bq/m$^3$] | Minimal value [Bq/m$^3$] | Average value [Bq/m$^3$] |
|---|---|---|---|
| I | 2218 | 32 | 131.5 |
| II | 2012 | 33 | 141.5 |
| III | 787 | 24 | 183.6 |
| IV | 1151 | 24 | 80 |

*Table 1. Radon concentration variations with seasons*

The lowest average value was in the IV quarter February-June, and the highest average concentrations were in III quarter October-February. Season changes directly depend on meteorological conditions –pressure, temperature, precipitation, level of ground and surface waters. In many houses, floors were made of ground and walls of Janja granite with increased contents of natural radionuclides.

The area of radioactive anomaly Stublovača was especially interesting. Local inhabitants build their houses of pyroclastic rocks of trachyte composition, causing the Rn concentrations to reach 8761 Bq/m$^3$ (in III quarter), total U contents of 43 g/t, Th 96 g/t and K 6.3 %. According to the content of natural radionuclides, radioactive anomaly Stublovaèa has thorium character. Although thoron ($^{220}$Rn) half-life is short (55 sec), it represents significant cause of internal radiation burden.

Atmogeochemical map of Rn concentration indoors was made in accordance with the recommendations of the World Health Organization (WHO), International Commission for Radiation Protection (ICRP), UN Commission for Atomic Energy Research (UNCEAR) and other international and national institutions.

According to these recommendations, total gamma radiation index of natural radioactive elements uranium, thorium and potassium should not exceed 1 ( 0.7 mSv per year). Intervention level of radon concentration indoors should not exceed values of 400 Bq/m$^3$ in old and 200 Bq/m$^3$ in new buildings.

Another, Bukulja ore region, was selected for hydrogeochemical profiling of wells and boreholes, based on geological, structural and metallogenetic characteristics. This ore region is built of the Palaeozoic rocks, as the oldest in the area, such as: phyllites, sericite and albite-chlorite-muscovite schists, metamorphosed sandstones, quartzites and marbles. Granitoides, as barriers of numerous uranium mineralizations, ore occurrences and deposits, were intruded in these rocks.

Contents of natural radionuclides in water and soil samples obtained from Bukulja region profiling are given in table 2.

| Radionuclide | water | | soil |
|---|---|---|---|
| Utot | (0.1 – 0,8) mg/l | avg*. 0.399 mg/l | (2.07 t- 10.61) ppm, avg. 3.945ppm |
| Ra | (0.04-0.3) Bq/l | avg. 0.06 Bq/l | N/A** |
| Rn | (5-1153) Bq/l | avg. 131.4 Bq/l | N/A |
| Th | N/A | | (1.35-24.12) ppm, avg. 13.8 ppm |
| K | N/A | | (0.33 - 7.06) %, avg. 3.5 %. |

*avg=average
** N/A = below the lower limit of detection

*Table 2. Concentrations of natural radionuclides in Bukulja region samples*

Low contents of U and Ra and extremely high Rn concentrations in waters were detected. Radon is not only the indicator of hidden ore bodies but also of deep fault structures – bearers of uranium mineralizations. It should be emphasized that in smaller settlements where inhabitants are supplied with water from wells and sources and take in radioactive content by ingestion, influencing the internal radiation burden (IRB). There are many quarries in the sampling area, and the stones from them are often used for decoration in the houses, with activity index higher than 1. The most important are: Glišića Majdan (i=169) and Gerasimov Majdan with activity index from 1.51 to 1.93[5-7].

Histogram of radon concemtrations in the water samples from wells is given at Figure 1.

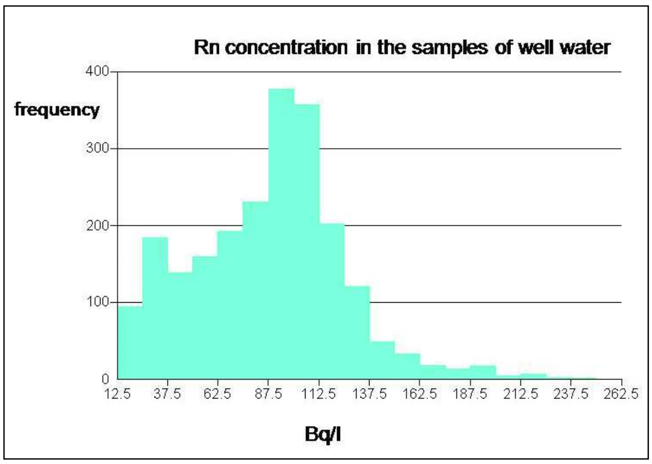


*Figure1. Radon concentrations in well waters*

Geopathological influence of radon from the same samples and the same area is given at Figure 2.

## CONCLUSION

The highest concentrations of radon in the houses were measured in the Gornja Stubla in vicinity of former uranium mines Gabrovnica, Mezdreja and Stara Kalna. Radon concentrations in them were in direct connection with the building material and geological basis. In this case, Janja granites were the source of radon and trachytes of Stublovaèa were the source of thoron. Increased concentration of radon in the waters (sources, wells and boreholes) in the area of Bukulja granitoide massif, as bearer of uranium mineralization is in direct dependence of structural-geological characteristics of exploration area.

Radon presence as direct indicator of uranium mineralization in different milieus of geosphere, defines the field of external and internal radiation burden of human population. Anomalous concentrations of radon were presented on geochemical map in the air of the housings and also the results of the measurements of U, Ra and Rn in waters, and U, Th and K in the rock samples from quarries and from Bukulja massif showed a tendency to anomalous values.

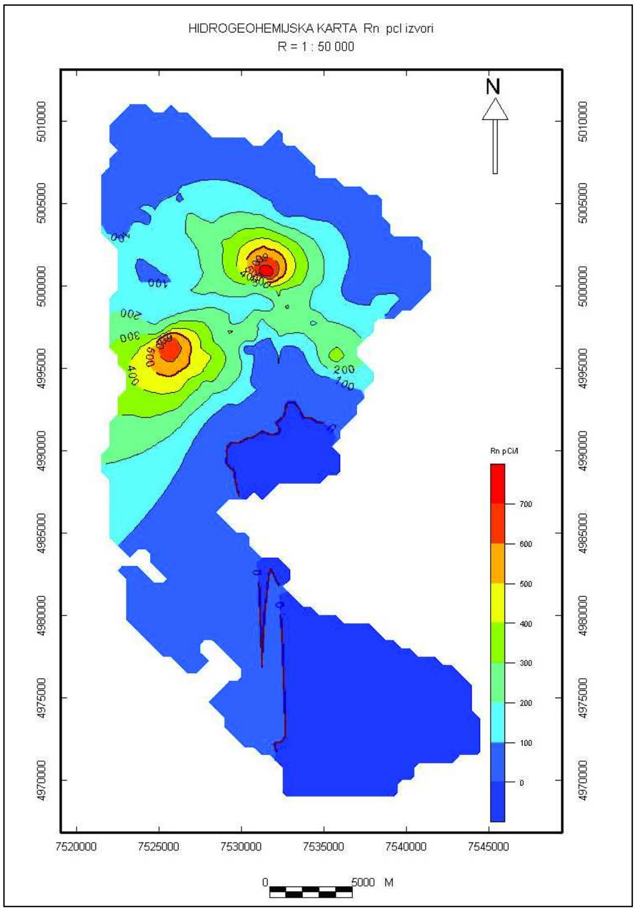


*Figure 2. Geopathological influence of Rn*

With additional biogeochemical, metallometric, geological-structural and other investigations we can single out the areas of radiogeochemical endemics. Characteristics of such areas are high radon concentrations in different milieus. It is geochemical indicator of the cause of ionizing radiation in rural environment. Geopathogenic zones for further medical investigations and prevention can be singled out through additional research and definition of radioecological areals in adequate geological-structural and geochemical milieus.

*Sažetak:*

Identifikacija geopatogenih zona se bazira na podacima dobijenim iz regionalnih istraživanja nuklearnih materijala u različitim fazama prospekcije geohemijskih područja. Podaci se odnose na koncentracije U, Th, K u tlu i stenama kao i na koncentracije U, Ra, Rn u vodi i Rn u vazduhu. Geohemijske karte zasnovane na geostrukturnim karakteristikama terena omogućavaju evaluaciju eksternog (ERB) i internog (IRB) radijacionog opterećenja tela. Prikazani rezultati za radiohemijska endemska istraživanja (RGHE) mogućih patogenih zona odnose se na geografsko područje Karpatsko - Balkanskih zemalja (Srbija i Makedonija).

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