Uranium content in ground water in Stara Planina Triassic sediments

Zoran Nikić[1], Jovan Kovačević[2], Branislav Radošević[3]

[1]Forestry Faculty, Kneza Višeslava 1, 11030 Belgrade, Yugoslavia.
[2]Geoinstitut, Rovinjska 12, 11000 Belgrade, Yugoslavia.
[3]"Advanced Systems", Vojislava Ilića 18, 11000 Belgrade, Yugoslavia.

Abstract. Uranium investigations in the Stara Planiana Triassic sediments were carried out for several decades. Some of these geological and hydrogeological results that are related to an established uranium deposit in the Dojkinci-Brlog-Jelovica-Ponor area are presented in this paper. Hydrogeological investigations were performed to investigate the chemical properties of ground water (e.g. anion-cation composition, microelement content, radioactive element content, pH value, Eh value). On the basis of these data a hydrogeological model was established for ground waters with increased uranium content.

Introduction

The Stara Planina massive, within the Carpathian-Balkan arch, is partly situated in western part of Bulgaria and partly in the easternmost part of Serbia.

In the southeast part of Stara Planina, belonging to Serbia, a uranium deposit is found, as well as several occurrencers. During several years of exploration numerous data were collected and many geological and hydrogeological problems were solved, helping to iterpret more precisely the increased uranium contens in ground waters of the Dojkinci area (N.N. *et al* 1985). The performed investigations covered an area of about 55 km^2.

Most of this area is uninhabited. The only settlements are Dojkinci, Jelovica, Rosomača and Senokos, which are connected by asphalt road to Pirot, the industrial and cultural centre of the area. The main occupation of the inhabitants is cattle breeding and agriculture to a lesser extent.

The main features of the investigated area

The area is featured by prominent mountain relief, ranging from 800 m to over 2000 m ASL (above sea level). The highest peak of this part of Stara Planina is Midžor (2169 m) with several others over 1700 m ASL (e.g. Crni Vrh: 1831 m, Kopren: 1935 m, Bratkova Strana: 1934 m, Tri Čuke: 1937 m).

The whole area has typical mountain climate, with sharp winters and a short, relatively warm, summer period. The snow often holds until mid May. The average annual rainfall for the period 1960-1990 ranges from 712.2 mm (Kamenica) to 981.1 mm (Dojkinci). Uneven distribution of rainfall is prominent, where much higher rainfall is represented in mountainous parts than in the valley parts of the area.

The significant streams in the area are the Kamenička, Rosomačka, Jelovička, Dojkinačka, and Toplodolska rivers. During their flow from the mountain heights these streams encounter a zone of karstified limestones with numerous sinks in the riverbeds. During the recession period, with low proticaj, all drained water sinks, so that down stream of the karstified limestones Kamenička, Rosomačka and Dojkinačka rivers are dry for several weeks during the year.

Geological composition and tectonic fabric of the investigated area

Stara Planina is a part of the Carpathian-Balkan geotectonic unit, with a strike of NW-SE through eastern Serbia, continuing to the north in Romania and to Bulgaria in the southeast (N.N. *et al* 1977).

The south-eastern part of Stara Planina, which is the subject of this work, is composed of quartz-albite-muscovite-chlorite schists of Riphean-Cambrian (R, Cm) age (R, Cm) with intrusions of granodiorite (δγ) and granodiorite-porphyrite (δγη). This schistose complex composes a wide belt on the crest of Stara Planina, with a supposed thickness of over 600 m (Fig. 1).

Transgressively over the crystalline schists lies the clastic complex of Lower Triassic (T_1), where sandstones and conglomerates are predominant rocks. Lower Triassic sediments are developed in form of a continuing narrow belt which strikes from Rudinja village in the northwest, and over the Kopren mountain towards the Yugoslav-Bulgarian border. Within this formation, in the area of Dojikinci and Jelovica, the lower member “Kopren” and the upper member “Dojkinci”, composed of varicoloured sandstones, are identified (Fig. 1).

The “Kopren” member is mostly composed of coarse grained quartzitic sandstones and red conglomerates, showing distinct cross-bedding. The thickness of this member is 200—250 m. Above the sediments of the “Kopren” member, conformably lies the varicoloured sandstones of the “Dojkinci” member. The varicoloured sandstone series is composed of dark reddish sandstones, greenish sandstones, siltstones and intra-formational conglomerates. The thickness of this member is 120—200 m.

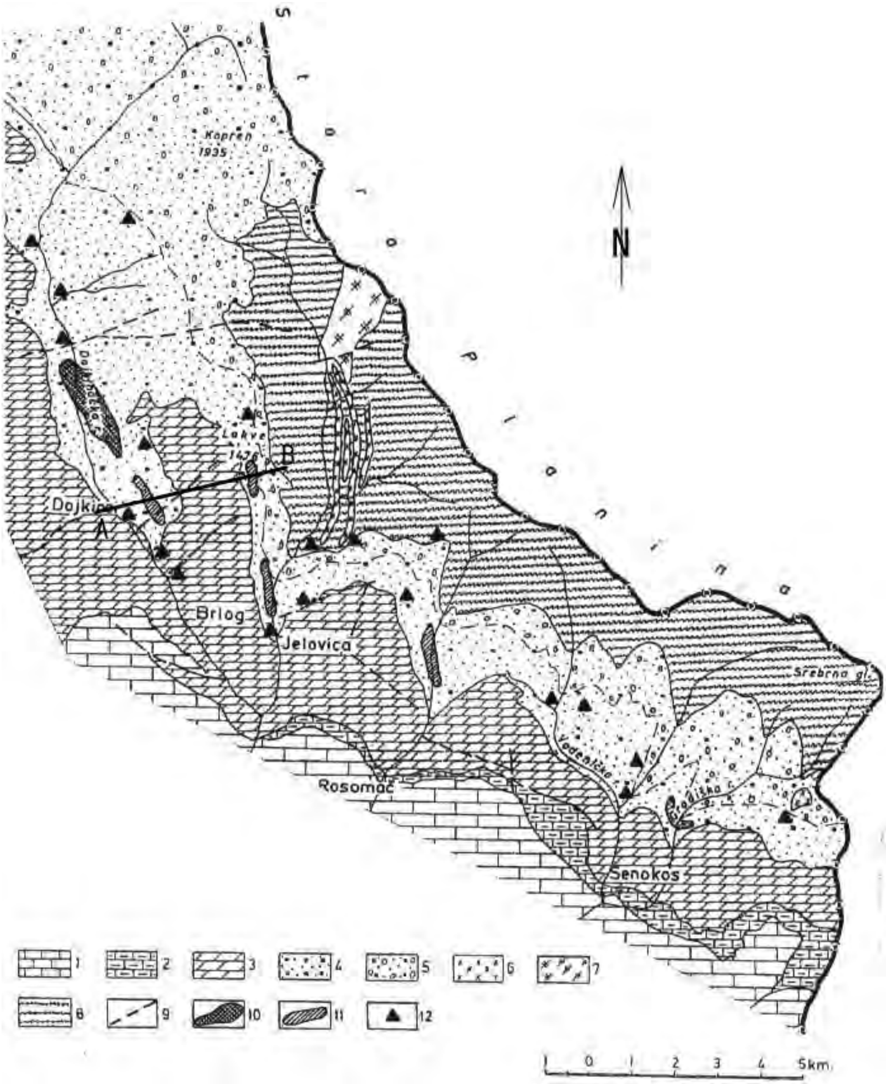

Fig. 1. Geological map of the investigated area. 1. Jurassic sediments, 2. Limestones sandstones and claystones (T,J), 3. Limestones, dolomitic limestones and dolomites, 4. Varicoloured sandstones (T_1), 5. Quartz sandstones and conglomerates (T_1), 6. Granodiorite- porphyrites, 7. Gabbro, 8. Quartz-albite-muscovite-chlorite schists (R, Cm), 9. Fault, 10. Uranium deposit, 11. Uranium occurrence, 12. Minor uranium mineralisation .

In the lower level of the varicoloured sandstones a geochemical barrier is identified with a strike NW-SE, up to 200 m wide and 5—25 m thick, representing the uranium bearing horizon (Kovačević 1997). The main feature of the uranium horizon is intensive alternation of sediments, reflected in changes of lithological composition, granulation, porosity, colour and presence of organic matter, sulphides and uranium mineralisation.

Over the "Dojkinci" member, lies a limestone and dolomite complex of Middle Triassic (T_2) age. The feature of this complex is alternation of thick beds of pure and karstified limestones with dolomitic limestones, dolomites and knotty limestones, somewhat less karstified. The thickness of this complex is about 250 m.

Outside the investigated area, upwards in the geological column of the Stara Planina anticline, there are flysch-like sediments of Jurassic and Lower Cretaceous age.

All the mentioned series have a mild dip 5—20°, rarely higher, and are generally inclined to the southwest.

Hydrogeological features of the investigated area

Important for this work is the existence of porous and permeable rock complexes, which are at the base limited by impermeable rocks.

The complex of limestones and dolomites of Middle Triassic (T_2) is identified as a hydrogeological area with aquifers of dissolutional and dissolutional-fracture porosity. The degree of their karstification is indicated by numerous typical karst occurrences and forms (e.g. sinks, caves, springs, caverns, sinkholes). In the period of high waters, complete underground rivers flow through caverns, with water yields of over 3 m^3/s for some springs (Veliko Jelovičko spring).

The sediments of Lower Triassic (T_1) age belong to the hydrogeological category of aquifers with fracture porosity. The intensity of their fracturing and existence of hydraulically connected fracture systems is indicated by the large number of permanent springs, with minimal water yields of 1—3 l/s.

The facies of greenschists of Riphean-Cambrian (R, Cm) age belongs to the hydrogeological category of more or less waterless rocks. This rock complex has a significant function of base impermeability and a lateral hydrogeological barrier for ground water in fractured Lower Triassic sediments.

Ground waters in the Dojkinci and Jelovica areas are according to their chemical composition hydrocarbonate, rarely hydrocarbonate-chloride and hydrocarbonate-sulphate, and very rarely hydrocarbonate-sulphate-chloride, sulphate-hydrocarbonate-choride and purely sulphate. By their cation composition they are mostly calcium, calcium-magnesium and rarely magnesium-calcium. The hydrocarbonate types are represented in the whole of the investigated area, hydrocarbonate-chloride types are found in the northern part in chlorite schists, while hydrocarbonate-sulphate and purely sulphate types are found in quartz conglomerates and conglomeratic sandstones north of Jelovica.

The content of radioactive elements (U, Ra, Rn) in ground waters of the Dojkinci-Jelovica area are found in the following ranges: uranium 0.1—40.9 μg/l, radium, 0.036—0.294 Bq/l and radon 0.52—63.2 Bq/l.

The mean content of uranium in ground water of the investigated area is 0.68 μg/l. The analysis of data, from 100 water samples, on normal dispersion and uranium and radium content range, in ground water of the investigated area, show that the increased uranium content is found in about 38%, and radium in about 34% of the water samples. The anomaly coefficient for uranium varies from 2.01 to 25.71, while for radium it varies from 1.0 to 8.2. Increased uranium contents were found in the area of Dojkinci-Paleški Vrh, in the area of Brlog-Jelovica and north of Ponor.

In the Dojkinci-Peleški Vrh, the increased content of uranium is found in ground water genetically connected with the varicoloured sandstones (0.3—6.8 μg/l). The anomaly coefficient varies from 1.85 to 15.22. The redox potential (Eh) of these waters ranges from +130 to 135 mV, indicating the transitional oxide-reduction environment with a pH value of 7.1—7.5, representing a neutral to mildly alkaline environment. According to the chemical composition, these waters belong to the hydrocarbonate-calcium type.

In the Brlog-Jelovica area several springs with an increased uranium content (1.9—40.9 μg/l) were found, situated in conglomerates, varicoloured sandstones and Granodiorite-porphyrites. The anomaly coefficient of uranium in these waters ranges from 2.01 to 25.71. Their redox potential varies from +125 to +135mV, indicating the transitional oxidational-reductional environment, while the pH value from 6.9 to 7.5 points to neutral to mildly alkaline waters. According to the chemical composition, these waters mostly belong to the hydrocarbonate-calcium type, with occurrences of hydrocarbonate-magnesium-calcium types. In the vicinity of Jelovica, the ground waters from the varicoloured sandstones and quartz conglomeratic sandstones are, according to their chemical composition, of mixed type, where hydrocarbonate-sulphate-calcium and purely calcium types dominate.

North of Ponor, in some ground waters within chlorite schists, increased contents of uranium were found, ranging from 1.5 to 20.4 μg/l, while their coefficients vary from 2.01 to 3.21. The Eh values of these waters range from 5.7 to 6.3, meaning that these waters are mildly acidic, indicating transitional oxidation-reduction environment. According to the chemical composition these waters are of the hydrocarbonate-chloride-calcium and sulphate-hydrocarbonate-chloride-magnesium type.

The increased content of radium in the investigated ground waters partly corresponds to the waters where increased contents of uranium were found. This mostly applies to the Dojkinci-Peleški Vrh area, and partly Jelovica area, while in the area north of Ponor increased contents of radium were not identified.

The SiO_2 content in the waters of the investigated area is generally low, ranging from 2 to 24 mg/l, with a mean value of 10 mg/l. This fact indicates that most of the ground water in the investigated area is in the zone of filtration, meaning that they are of surface origin and there is a fast percolation of water (Nikić 2001).

Hydrogeological model of ground water formation with increased uranium content in Dojkinci-Jelovica area

The general characteristics and some particularities presented earlier represent a base for understanding and interpreting the role of hydrogeological conditions in the Dojkinci area and formation of ground water with an increased content of uranium (Fig. 2).

Due to favourable climate, hydrogeological and morphological conditions of charge of the karst aquifer in the Middle Triassic limestones (T_2) is done by infiltration of rain and snow water and sinking of surface water courses, flowing from surrounding non-karstic areas. Drainage of such an aquifer is done partly through springs and partly by sinking downwards. In the waters of karst springs an increased uranium content was not identified.

Charging of the fracture aquifer, formed in Lower Triassic sediments (T_1), is done by infiltration of rain and snow water, filtration of ground water from the limestones in the base, and surface water courses which flow from the surrounding impermeable areas. In the surface, the outcrops of Lower Triassic sediments, represent the areas of direct charge to the aquifer, while the indirect charge is from the karst aquifer which is in the base.

By filtration along hydraulically connected fissure systems, the water comes in the contact with the uranium bearing horizon. By contact of water with uranium ore, new uranium compounds are formed, contaminating the ground water transported into favourable geochemical conditions. Migrativity of uranium in ground water of Dojkinci-Jelovica area, varies and depends on chemical properties of ground waters, controlled by geological composition. The chemical composition of waters with a high HCO_3 content is mildly alkaline and mostly in an oxidation environment, indicating that these ground waters represent an aggressive environment collecting and maintaining uranium in form of complex alkaline uranilo-carbonate. The coefficient of uranium migration in the waters of hydrocarboneta types varies from 0.012 to 0.448, with a mean value of 0.145. In hydrocarbonate-magnesium calcium waters the migration coefficient ranges from 0.013 to 0.101, with a mean value of 0.061. The hydrocarbonate-sulphate-calcium waters have a uranium migration coefficient ranging from 0.096 to 1.363, with a mean value of 0.330. The hydrocarbonate-sulphate-calcium waters and hydrocarbonate-chloride-calcium-magnesium waters have an increased uranium migration coefficient ranging from 0.112 to 1.482, with a mean value of 0.769.

Due to the higher position of ground water within the Lower Triassic aquifer, in relation to the local erosional base, represented by the Dojkinačka Reka river bed, favourable dip of hydrogeological base and the aquifer in relation to the erosional base, plentiful quantities of infiltrational waters and the existence of hydraulically connected fissure systems, the process of gravitational discharge of ground water, enriched by uranium, is done dosed through some springs. The locations of these springs are determined by the distribution of fault systems, fractures, and fissures that are found on the surface. For the motion of ground water within the aquifer and their spontaneous discharge on the surface, the influence of distribution of water energy in space (mechanical energy) in the form of potential energy is of importantance.

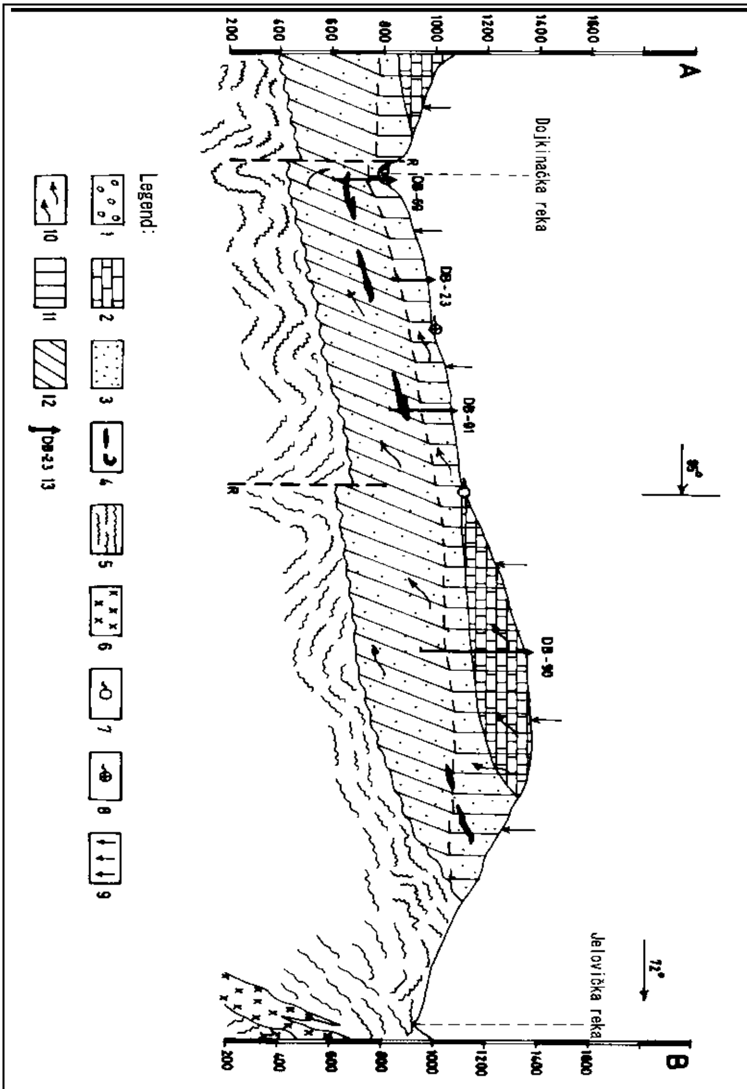

Fig. 2. Thematic hydrogeological model of formation of ground water with increased uranium content in Dojkinci-Jelovica area. 1. Alluvial deposits, 2. Limestones and dolomites, 3. Sandstones and conglomerates, 4. Uranium mineralization, 5. Schists, 6. Granodiorites, 7. Karst spring, 8. Spring. Water with increased uranium content, 9. Atmospheric precipitation, 10. General direction of ground water flow, 11. Zone of faster filtration (water exchange), 12. Zone of slower filtration, 13. Exploratory drill hole.

Impact on groundwater from radionuclide emission

Discharge of water from the Lower Triassic sediments is done mainly through fracture systems of largest dimensions, where the spring yield is the highest, while at the end of the recession period, during the lowering of the ground water level, the water is discharged from even smallest fissure systems, with distinguished participation of water from the lower part of the aquifer. In that way, together with ground water discharged along fracture systems, the uranium comes to the surface in form of solution.

Conclusion

The variety of hydrochemical features of ground waters indicates their genetic bond with particular rocks with which they were in contact during their flow along connected fracture systems. Increased content of radioactive elements in ground waters from some drill holes and springs, points to a conclusion that it is a consequence of their direct contact with uraniferous horizons formed in Lower Triassic sediments. The factors controlling the occurrence of increased uranium contents in ground waters of Stara Planina, in the vicinity of Dojkinci-Brlog-Jelovica-Ponor, are of geological, hydrogeological, tectonic, climate, and geomorphological type and at the same time in simultaneous interaction.

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