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HYDROGEOCHEMICAL INVESTIGATION OF GROUND AND SURFACE WATER IN THE NEOGENE - QUATERNARY SEDIMENTS**

Abstract

Geochemical investigations in order to identify the mineral deposits, as well as the sources of water supply of rural settlements included in the area of Bela Crkva, between the Nera and the Karaš rivers, draining the Neogene and Quaternary sediments. During the prospecting, water samples were collected from surface water streams, springs, wells, drills, water reservoirs, as well as the samples of stream sediments and rocks. Hydrogeochemical investigations are of significance, where, in the water of Vrsac hills, elevated levels of radionuclides U, Ra and Rn, and other toxic elements were detected with values above maximum allowed concentrations for drinking water. The wells of rural households had increased levels of Fe and Mn, as well as the increased mineralization, conductivity, and nitrogen cycle. The concentration of heavy metals in water wells was increased in the most settlements around Bela Crkva, as well as in Češko Selo, Banatska Subotica, Kuštilj, Jablanka and Karaš river.

From the water accumulations formed in the Quaternary sediments southwest from Bela Crkva, samples of water, mud, overbank sediments, and A-horizon were collected. The water samples were determined on: Na, K, Ca, Mg, Fe, Mn, Al, NH_4 , NO_3 , SO_4 , HCO_3 , Cl, F, Ep, Eh, pH value, mineralization, Pb, Cd, Co, Ni, Cr, Cu, Zn, H_2S , O_2 , CO_2 , U, Ra, Rn, As, Hg, Br. In the solid samples, Au, Ag, Pb, Zn, and other elements, as well as the content of ^{238}U , ^{232}Th and ^{40}K were determined. Test results are presented in tables, diagrams, and hydrogeochemical maps.

Keywords: hydrogeochemistry, prospecting, radioactivity, toxic elements, Neogene and Quaternary sediments.

INTRODUCTION

The presence of uranium deposits in Romania (Čudanovica Dobre), located in the zone of Rešica-Moldava Nova, relatively close to the border with Serbia im-

posed the need for research in Vršac-Bela Crkva region, as a perspective area for discovering uranium deposits and other mineral materials (Figure 1).

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** This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia (Project OI 176018: "Geological and Ecotoxicological Research in Identification of Geopathogenic Zones of Toxic Elements in Drinking Water Reservoirs – Analysis of Methods and Procedures for Reduction the Effects of Biogeochemical Anomalies")

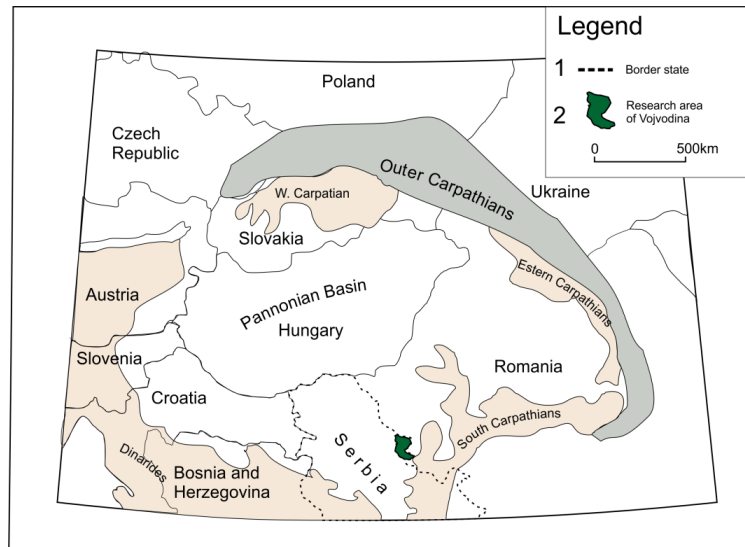


Figure 1 Location of research area of the Pannonian Basin in Serbia

The earliest studies dating back to the 19th century (1880-1886) [1], when the first geological map of the Vršac area, scale 1:144000, with explanation was made. The most important data of studied area were gathered during research of oil and gas which were synthesized and then used for creation of book interpreter for the map of Bela Crkva 1:100000 [2, 3]. Researches included numerous geological, hydrogeological, geophysical, geochemical investigations. For the water supplying of Bela Crkva and surrounding rural settlements, in the area of Bela Crkva, the systematic monitoring of chemical elements distribution in different environments of geosphere was carried out, which is of special significance in detecting the natural and anthropogenic influence on living environment, and for defining the ecological status as well [4]. The largest number of data was collected during the regional research of nuclear materials in the area of Vršač hills - Bela Crkva [5], during the preparation of maps geochemical-ecological atlas 1:50000 [6]. The area of Bela Crkva is located in the southeastern Banat and presents the southeast end of the

Pannonian plain. In morphogenic terms, it is divided into three parts: sandstone west of the Bela Crkva, loess plateau and river lake terrace while the lowest parts of terrain are presented as the alluvial plains. An aquifer is formed in them with free level or so called the first aquifer, which is locally exploited through shallow wells for water supply or irrigation.

METHODOLOGY

Lithochemical, metalometric and hydrogeochemical researches of the area of Bela Crkva (Figure 1) were applied. Sampling network was adjusted to the morphological and hydrographic characteristics of the terrain. The surface flows and shallow and deeper aquifers were tested.

In all water samples, contents of U, Eh, pH and Ec were determined. In water with the increased content of uranium, and pH, Eh and Ec changes, the complete chemical analyses were conducted: anion-cation composition, content of microelements and radioactive elements U, Ra and Rn, content of gases O₂, CO₂, H₂S (Figure 2).

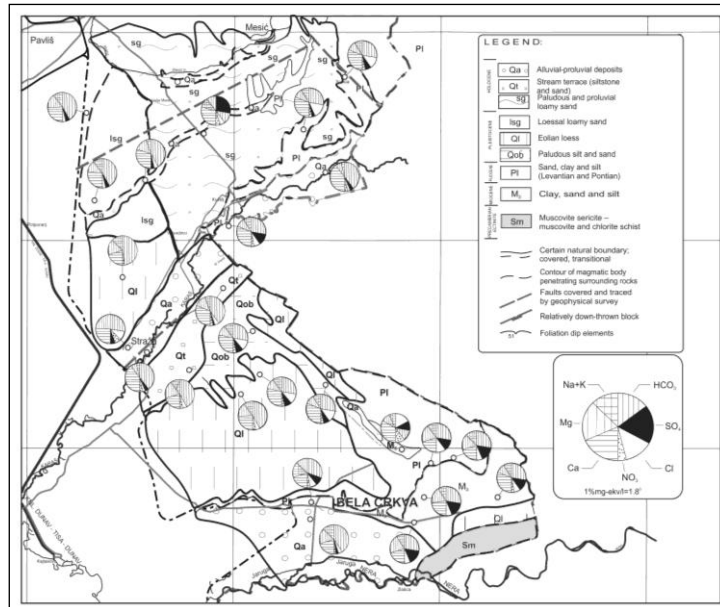


Figure 2 Anion-Cation composition of water samples from the area of Bela Crkva

For determining the metal content in water, Atomic Absorption Spectrophotometry was applied (AAS Perkin Elmer M-306). For other components in water solutions, colorimetric, volumetric, potentiometric and turbidometric methods were used. Uranium was determined using the laser fluorimeter UA of Canadian production of Scintrex company, with fluran as the characteristic reagent. Radium concentrations were measured by radon detector RD-200 EDA, and radon concentrations were measured by radon emanometer ETR-1 Scintrex, Canada.

RESULTS AND DISCUSSION

The quaternary formations of Pleistocene and Holocene cover about 70% of total area included within regional research of uranium in the area Vršac - Bela Crkva. The terrain is made of Quaternary formations of fluvial and aeolian origin which were deposited at the time when Pannonian Basin became mainland, with rivers, lakes and puddles.

The formations are presented as puddle alevrolites (Qab), aeolian loess (Ql), and loess clays (Figure 2). These sediments were created in continental conditions, and their origin is fluvial and aeolian. The thickness of Pleistocene in the Vršac area is about 30-60 m, and in the northern and western part reaches 100 m. Holocene is developed in morphologically the lowest parts of the terrain (north and south from the Vršac hills) and along the beds of smaller or bigger rivers Nera and Karaš. The sediments have a heterogeneous composition and consist of: proluvial suglines (Sg), debris of river terraces (alevrolites and Qz sand) and alluvial-proluvial creations (bed facies, floods with gravels, sand and alevrolites - Qa).

In hydrogeological terms, the terrain has a complex geological structure in the northern part from Bela Crkva (Vršac hills) the terrain is composed of old rock masses: granites, gneisses, albite-muscovite schists, and on the other side of Bela Crkva area the terrain is composed of a thick strata of the Tertiary and Quaternary sediments. The position of rock masses and the character of

porosity in them caused formation of different types of aquifers in that part of the terrain. In terms of structure, the structural elements are significant which extend in the NE-SW direction, and in that direction, the surface flows that drain this area are developed. In deeper parts of the terrain there are no accumulations of free underground water, so the faults which intersect the terrain do not have higher hydrogeological significance.

The underground water is mainly $\text{HCO}_3\text{-Ca-Mg}$ type, with low mineralization (about 0,4-0,5 g/L), pH neutral and moderately hard; it is contained in an open hydrogeological structure, where there is an intense water exchange, so the contents of certain chemical components are subjected to the time changes. Free aquifer of compact type was formed on the entire expanse of alluvial-lake sediments. This aquifer was developed in the alluvial plain of the rivers Karaš and Nera, then in the sand which is locally contained at different depth [7]. Depth to groundwater is mainly 5-10 m, and on smaller expanse depth varies above and beneath this value. Hydroisopleths of the first aquifer mainly follow the isopleths of the terrain and point to the directions of underground water drainage that is towards the erosive basin of

the rivers Nera and Karaš. In the area of Bela Crkva and wider (Vršac hills) in the phase of hydrogeochemical prospection, the samples of water were collected from surface flows, spring wells and lakes. According to the anioncation composition, water belongs to the hydrocarbonate type (Figure 2).

According to the presence of cations, calcium and magnesium prevail in water, which classifies it as calc-magnesium (Ca-Mg), and magnesium-calcium (Mg-Ca) type of water. Among other cations, Na^+ K^+ are present but in considerably smaller quantity. Mineralization varies within the interval of 138 mg/L - 4557 mg/L in Vršac hills. The concentration of hydrogen ions varies from 5.5 to 7.9 so this water can be classified as weakly acidic to slightly alkaline. The redox potential (Eh) was determined in every water sample. On the basis of Eh values, we can conclude that water is mostly located in oxidizing conditions. The Eh values are within the interval from -65 mV to +190 mV.

Contents of microelements vary in the interval for: Mo from 1-14 ppb, \approx 2ppb, Li from 3-60 ppb, \approx 5 ppb, Sr from 30 - 3250 ppb, \approx 300 ppb, Zn from 3 - 3800 ppb, \approx 13 ppb (Figure 3).

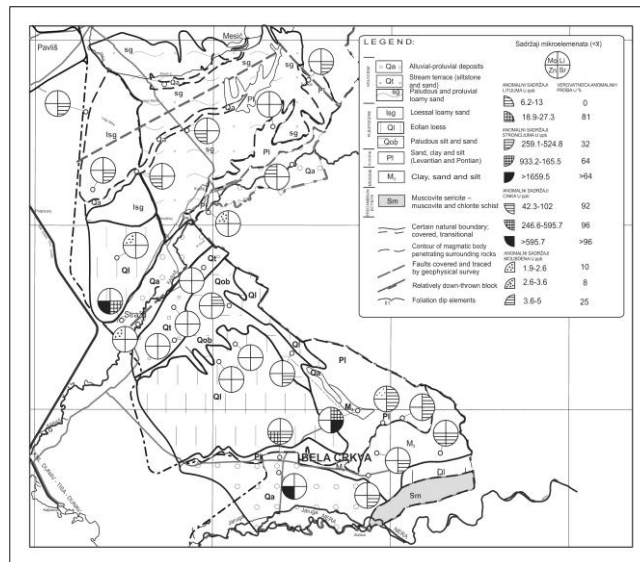


Figure 3 Hydrogeochemical map of microelements content

The increased contents of Li are caused by the vicinity of granite intrusion in Vršac hills, zone of fine grained gneisses with leptinolites, aplites and strings of pegmatite. Clay materials in the sediments of Bela Crkva, unlike other alkaline elements (Lithium, Rubidium, Cesium) absorb weakly Sr. Since its ion has a large radius, it cannot be absorbed into compact structure of clay minerals with the exception of multistrata structure of monmorionites. The increased content of Sr is related to hydrocarbonate water which probably originate from granite intrusion. Anomalous concentrations of U, Ra and Rn in water can be found in the Vršac hills area.

In the Bela Crkva area, including water accumulations of lakes: Vračev Gaj, Šaran lake, Bager lake, Veliko lake, the water analysis showed that water has slightly alkaline character. Concentration of ammonium ion, according to maximum allowed concentration (MAC) of dangerous substances in

water [8], has classified this water in the III/IV category and water from the lakes is classified in the I and II category. The nitrogen cycle is intense but content of nitrates does not exceed the MAC value. None of the samples detected hydrogen-sulfide as a product of anaerobic decomposition, which means that there is no process of rotting and decay. This water by its composition is calcium-sulphate, and by mineralization mainly belongs to water of the I category that is the II category [9]. It is important to point out that this water by the content of heavy metals can be classified as water of the I/II category according to the mentioned regulation.

The increased content of ammonia and nitrates gives a bad bacteriological image. In most wells there is an increased content of Fe and Mn which is a characteristic of underground water in this area, so water from certain wells shown in the hydrogeochemical map (Figure 4) cannot be used for drinking [10].

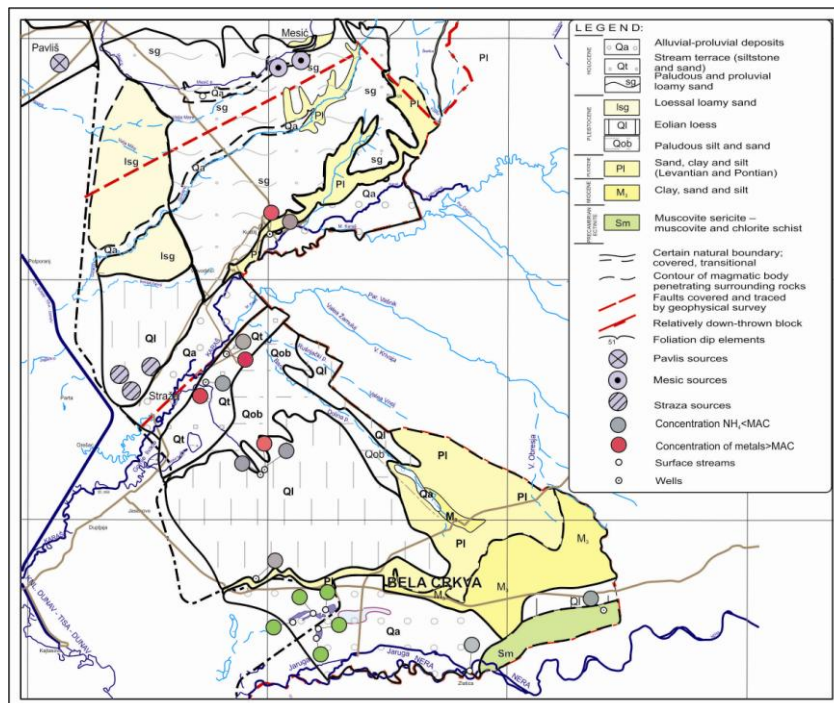


Figure 4 Geocological map of chemical content above maximum allowable concentration (MAC)

CONCLUSION

In the phase of regional semi-detail and detail researches in the region of Bela Crkva and Vršac hills, the aquifer level was defined for water supply of rural settlements.

During the hydrogeochemical prospection, in every sample of water from surface flows, spring wells, and borings, complete chemical analyses were performed, which are relevant for researching the mineral materials deposits, and they present a base for the assessment of the influence of geological composition on the living environment. In the area of Bela Crkva, anomalous concentrations of elements in water of rivers Karaš and Nera and other water points are evident and they are a consequence of anthropogenic influence of excessive pollution from which the geoecological maps were done.

Presented results of chemical analyses of water, aquifer type and water quality of selected water points in rural settlements, represent a basis in spatial planning and living environment protection.

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HIDROGEOHEMIJSKA ISPITIVANJA KVALITETA PODZEMNIH I POVRŠINSKIH VODA U NEOGENIM – KVARTARNIM SEDIMENTIMA **

Izvod

Geološko-geohemijska istraživanja u cilju identifikacije ležišta mineralnih sirovina, kao i izvorišta za vodosnabdevanje ruralnih naselja obuhvatila su područje Bele Crkve, između reka Nere i Karaša, koje dreniraju sedimente Neogena i Kvartara. U toku prospekcije, prikupljeni su uzorci voda iz površinskih tokova, izvora, bunara, bušotina, vodenih akumulacija, potočnih sedimentata i uzorci stena. Značajna su hidrogeohemijska ispitivanja, u kojima su u vodama otkriveni povišeni sadržaji radionuklida U, Ra i Rn u području Vršackih brda, kao i drugih toksičnih elemenata, čije su vrednosti iznad maksimalno dozvoljenih u vodi za piće. U bunarima seoskih domaćinstava povećani su sadržaji Fe, Mn, a povećani su i mineralizacija, provodljivost i azotni ciklus. Koncentracija teških metala u bunarskim vodama je povećana u većini naselja oko Bele Crkve kao i u naseljima Češko selo, Banatska Subotica, Kuštilj, Jablanka kao i reka Karaš.

Od vodenih akumulacija nastalih u kvartarnim sedimentima JZ od Bele Crkve; prikupljeni su uzorci voda muljevite komponente s dna jezera, overbank sedimentata i A-horizonta. U uzorcima voda određivani su : Na, K, Ca, Mg, Fe, Mn, Al, NH₄, NO₃, SO₄, HCO₃, Cl, F, Ep, Eh, pH, mineralizacija, Pb, Cd, Co, Ni, Cr, Cu, Zn, H₂S, O₂, CO₂, U, Ra, Rn, As, Hg, Br, u čvrstim uzorcima Au, Ag, Pb, Zn, U i dr. elementi, kao i sadržaji U, Th i ⁴⁰K. Rezultati ispitivanja prikazani su: tabelama dijagramima i hidrogeohemijskim kartama.

***Cljučne reči:** hidrogeohemija, prospekcija, radioaktivnost, toksični elementi, sedimenti neogena i kvartara*

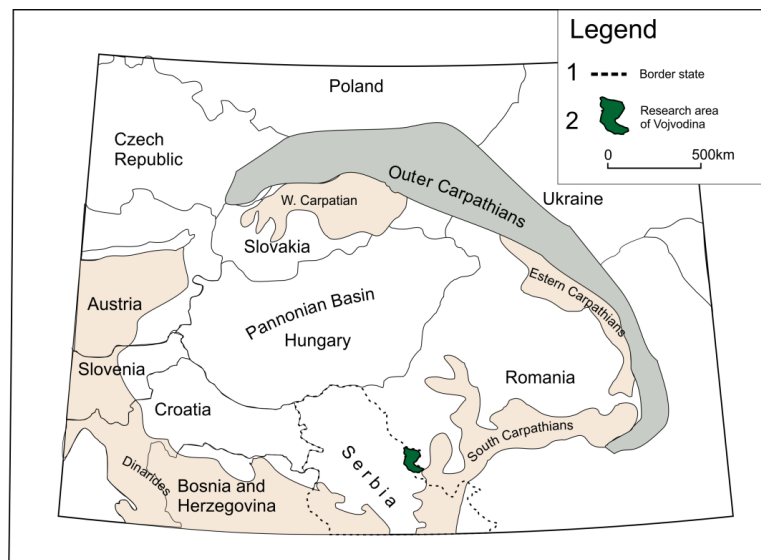
UVOD

Prisustvo uranskih ležišta u Rumuniji (Čudanovica Dobre), koja se nalaze u zoni Rešica - Moldava Nova, na relativno bliskom rastojanju od granice sa Srbijom,

nametnula su potrebu za istraživanjem regiona Vršac - Bela Crkva, kao perspektivnog područja za otkrivanja ležišta urana i drugih mineralnih sirovina (Slika 1).

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** Ovaj rad je realizovan uz podršku Ministarstva prosvete, nauke i tehnološkog razvoja Republike Srbije (projekat OI 176018: „Geološka i ekotoksikološka istraživanja u identifikaciji geopatogenih zona toksičnih elemenata u akumulacijama vode za piće– istraživanje metoda i postupaka smanjivanja uticaja biogeohemijskih anomalija“)



Sl. 1. Lokacija istraživanog područja Panonskog basena u Srbiji

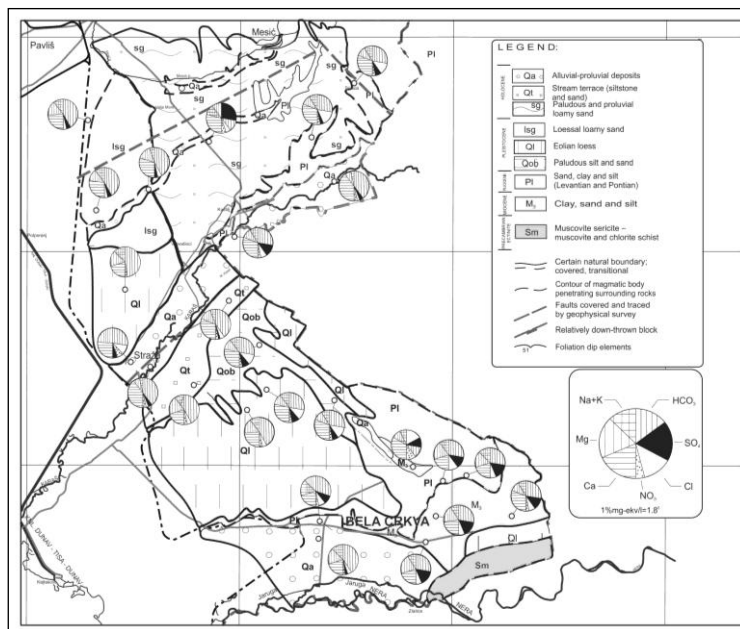
Najranija istraživanja datiraju iz vremena 19. veka (1880-1886. god.), kada je urađena prva geološka karta okoline Vršca u razmeri 1:144000 sa tumačem [1]. Najvažniji podaci o ispitivanom području dobijeni su prilikom istraživanja nafte i gasa, koji su korišćeni za izradu tumača za List Bela Crkva 1:100000 [2, 3]. Istraživanjima su obuhvaćena brojna ispitivanja iz oblasti geologije, hidrogeologije, geofizike, geochemije i dr. Za potrebe vodosnabdevanja Bele Crkve i okolnih ruralnih naselja, u ovom području Bele Crkve izvršeno je sistematsko praćenje distribucije hemijskih elemenata u različitim sredinama geosfere, što je od posebnog značaja za praćenje prirodnog i antropogenog uticaja na životnu sredinu i definisanje ekološkog statusa [4]. Najveći broj podataka prikupljen je za vreme regionalnih istraživanja nuklearnih sirovina na području Vršćkih bregova-Bele Crkve [5], odnosno tokom izrade geochemijskog-ekološkog atlasa 1:50000 [6]. Područje Bele Crkve nalazi se u jugoistočnom Banatu i predstavlja krajnji jugoistočni deo Panonske nizije. U morfološkom smislu podeljeno je na tri celine:

peščaru, zapadno od Bele Crkve, Lesnu zaravan i rečno jezersku terasu, a najniži delovi terena predstavljeni su aluvijalnim ravnima. U njima je formirana izdan sa slobodnim nivoom ili takozvana prva izdan, koja se lokalno eksploatiše preko plitkih kopanih bunara za vodosnabdevanje ili navodnjavanje.

METODOLOGIJA

Litogeochemijska, metalometrijska i hidrogeochemijska ispitivanja su primenjena u fazi regionalnih istraživanja na području Bele Crkve (Slika 1). Morfološkim i hidrografskim karakteristikama terena prilagođena je mreža uzorkovanja. Ispitivani su površinski tokovi plitke i dublje izdani.

U svim uzorcima voda određeni su sadržaji U i vrednosti Eh, pH i Ep. U vodama sa povišenim sadržajem urana, promena pH, Eh i Ep urađene su kompletne emijske analize voda, i to: anjonsko-kationski sastav vode, sadržaj mikro-elemenata i radioaktivnih elemenata U, Ra i Rn, sadržaj gasova O₂, CO₂, H₂S (Slika 2).



Sl. 2. Anjonsko-katjonski sastav uzoraka vode sa područja Bele Crkve

Za određivanje sadržaja metala u vodama, primenjena je Atomska Apsorpciona spektrofotometrija (AAS Perkin Elmer M-306). Za ostale komponente, u vodenim rastvorima korišćene su kolorimetrijske, volumetrijske, potencimetrijske i turbidimetrijske metode. Uran je određivan pomoću laserskog fluorimetra UA firme Scintrex kanadske proizvodnje, sa fluranom kao karakterističnim reagensom. Koncentracije radijuma su merene pomoću radon detektora RD-200 EDA, a koncentracije radona merene su emanometrom ETR-1 Scintrex, Kanada.

REZULTATI I DISKUSIJA

Kvartarne tvorevine Pleistocena i Holocena, zahvataju oko 70% ukupne površine obuhvaćene regionalnim ispitivanjima urana područja Vršac-Bela Crkva. Teren je izgrađen od kvartarnih tvorevina, koje su fluvijalnog i eolskog porekla i taložene u vreme kada je Panonski basen postao kopno, sa rekama, jezerima i barama.

Tvorevine su predstavljene barskim alevrolitima (Qab), eolskim lesom (Ql), lesoidnim glinama (Slika 2). Ovi sedimenti su stvoreni u kontinentalnim uslovima, a porekla su fluvijalnog i eolskog. Debljina pleistocena u području Vršca je oko 30-60 m, a u severnom i zapadnom delu dostiže 100 m. Holocen je razvijen u morfološki najnižim delovima terena (severno i južno od Vršačkih brda) kao i duž korita manjih ili većih reka Nere i Karaša.

Sedimenti su heterogenog sastava i sastoje se od: barskih, proluvijalnih suglina (Sg), drobina, rečnih terasa (alevriti i peskovi Qz) i aluvijalno-proluvijalnih tvorevina (facija korita, plavina sa šljunkovima, peskovima i alevrolitima-Qa).

U hidrogeološkom smislu teren je složene geološke građe. U severnom delu od Bele Crkve (Vršačkih brda) izgrađen je od starih stenskih masa – graniti, gnajsevi, albitsko-muskovitski škriljci a sa druge strane područje Bele Crkve debelih naslaga tercijarnih i kvartarnih sedimenata. Položaj stenskih masa i karakter poroznosti u njima

uslovlili su da se u tom delu terena formiraju izdani različitog tipa.

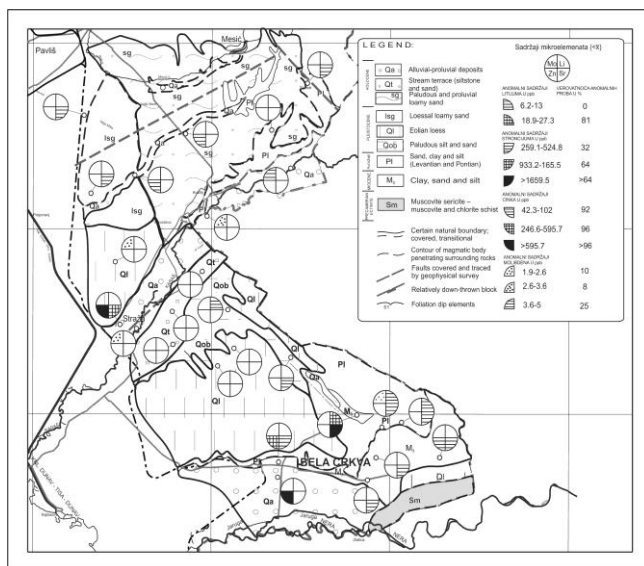
U strukturnom pogledu, značajni su strukturni elementi pravca pružanja SI-JZ, te su u tom smeru razvijeni površinski tokovi koji dreniraju ovo područje. U dubljim delovima terena nema akumulacija slobodnih podzemnih voda, pa ni rasedi koji presecaju teren nemaju veći hidrogeološki značaj.

Podzemne vode su pretežno $\text{HCO}_3\text{-Ca-Mg}$ tipa, slabomineralizovane (oko 0,4-0,5 g/L), neutralne reakcije i umereno tvrde, koje se nalaze u otvorenoj hidrogeološkoj strukturi, gde se vrši intenzivna vodozmena, pa su i sadržaji pojedinih hemijskih komponenti podložni promenama u funkciji vremena.

Slobodna izdan zbijenog tipa formirana je na celom prostranstvu aluvijalno-jezerskih sedimenata. Ova izdan je razvijena u aluvijalnoj ravni reke Karaš i Nera, zatim u peskovima koji se lokalno nalaze na različitoj dubini [7]. Dubina do podzemne vode je pretežno 5-10 m, a na manjem prostranstvu dubina varira iznad i ispod ove vrednosti. Hidroizohipse prve izdani uglavnom prate izohipse terena i ukazuju na pravce dreniranja podzemnih voda, odnosno ka erozionom bazisu reke Nera i Karaš.

U području Bele Crkve i šire (Vršačka Brda) u fazi hidrogeohemijske prospekcije, prikupljeni su uzorci voda iz površinskih tokova izvora bunara i jezera. Prema anjonsko-katjonskom sastavu, vode pripadaju hidrokarbonatnom tipu (Slika 2). Po zastupljenosti katjona, u vodama prevladavaju kalcijum i magnezijum, što ih svrstava u kalcijско-magnezijske (Ca-Mg), magnezijско-kalcijске (Mg-Ca) tipove voda. Od ostalih katjona i znatno manjoj količini su Na+K. Mineralizacija se kreće u intervalu (138 mg/L do 4557 mg/L). U Vršačkim brdima koncentracija vodonikovih jona varira od 5,5 - 7,9, pa se ove vode mogu svrstati u grupu slabokiselih do slaboalkalnih voda. Oksidaciono redukcionni potencijal (Eh) određivan je u svakom uzorku vode. Na osnovu vrednosti Eh možemo zaključiti da se vode nalaze pretežno u oksidacionim uslovima. Vrednosti Eh, nalaze se u intervalu od -65 mV do +190 mV.

Sadržaji mikroelemenata variraju u intervalu za: Mo od 1-14 ppb, Pb = 2 ppb, Li od 3-60 ppb, Cu = 5 ppb, Sr od 30-3250 ppb, Mn = 300 ppb, Zn od 3 ppb – 3800 ppb, Ni = 13 ppb (Slika 3).



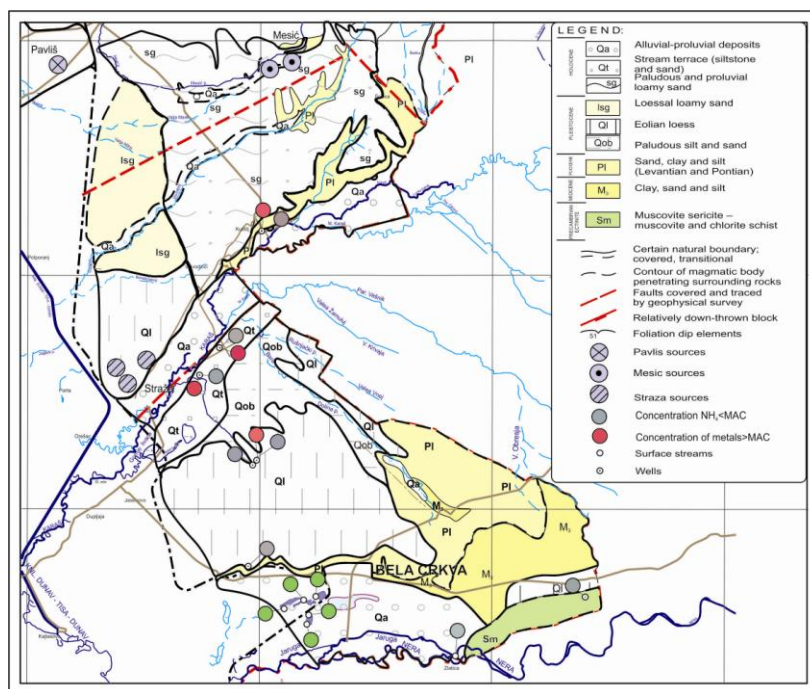
Sl. 3. Hidrogeohemijska karta sadržaja mikroelemenata

Povećani sadržaji Li uslovljeni su bližinom granitske intruzije u Vrščkim brdima, zone sitnozrnih gnajseva sa leptinolitima, aplitima i žicama pegmatita. Glinoviti materijali u sedimentima Bele Crkve, za razliku od drugih alkalnih elemenata (Litijum, Rubidijum, Cezijum) slabo apsorbuju Sr. Kako je njegov jon velikih razmera to ne može biti apsorbovan u zgusnutu strukturu glinovitih minerala, sa izuzetkom višeslojne strukture monmorionita. Povišeni sadržaji Sr vezani su za hidrokarbonatne vode i verovatno vode poreklo od granitskog intruziva. Anomalne koncentracije U, Ra i Rn u vodama nalaze se u području Vrščkih Brda.

U području Bele Crkve uključujući i vodene akumulacije jezera: Vračarev Gaj, Jezero, Šaransko jezero, Bagersko jezero i Veliko Jezero, analize voda su pokazale da vode imaju slabo alkalni karakter. Koncentracija amonijum jona, prema maksimalno

dozvoljenoj količini opasnih materija u vodama [8] svrstava ovu vodu u kategoriju III/IV, a vode iz Jezera u I i II kategoriju. Izražen je azotni ciklus ali sadržaj nitrata ne prelazi maksimalno dozvoljenu vrednost. Ni u jednom uzorku nije detektovan vodonik-sulfid, kao proizvod anaerobnog raspadanja, što znači da nema procesa truljenja i raspadanja. Ove vode su po svom sastavu uglavnom kalcijum-sulfatne, a po mineralizaciji uglavnom pripadaju vodama I odnosno II kategorije [9]. Bitno je istaći da se, i po sadržaju teških metala mogu svrstati u I/II kategoriju voda prema navedenoj uredbi.

Povećani sadržaji amonijaka i nitrata daju lošu bakteriološku sliku. U većini bunara je povećan sadržaj Fe i Mn što je karakteristika podzemnih voda ovog područja, pa se voda iz pojedinih bunara prikazanih na hidrogeohemijskoj karti (Slika 4) ne može koristiti kao voda za piće [10].



Sl. 4. Geokološka karta hemijskih sadržaja preko maksimalno dozvoljene koncentracije (MDK)

ZAKLJUČAK

U fazi regionalnih, poludetaljnih i detaljnih istraživanja, u region Bele Crkve i Vršaćkih brda, definisan je nivo izdani za vodosnabdevanje ruralnih naselja.

U toku hidrogeohemijske prospekcije, u svakom uzorku vode: površinskih tokova, izvora, bunara i bušotina, urađene su hemijske analize od značaja za istraživanje ležišta mineralnih sirovina, a koje čine osnovu u proceni uticaja geološkog sastava na životnu sredinu. U području Bele Crkve, evidentne su anomalne koncentracije elementa u vodama reke Karaš i Nere i drugih vodopunktova, koje su posledica antropogenog uticaja prekograničnog zagađenja, na osnovu čega su urađene geološke karte.

Prikazani rezultati hemijskih analiza voda, tip izdani i kvalitet voda izabranih vodopunktova u ruralnim naseljima čine osnovu u prostornom planiranju i zaštiti životne sredine.

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