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Natural radioactivity of groundwater in Serbia

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Abstract. Activity concentrations of radionuclides ^{40}K, ^{228}Ra, ^{226}Ra, ^{238}U and Th^{232} and gross alpha and beta activities were analyzed in more than 100 samples of groundwater in Serbia. The highest gross alpha activity was recorded at 1.33 Bq/L (average 0.12 Bq/L), while the highest beta activity was 5.43 Bq/L (average 0.68 Bq/L). The potassium isotope ^{40}K exhibited the highest active concentration (2.6 Bq/L) and was the largest contributor to the gross natural beta activity. Among the analyzed samples, 28 were found to have elevated beta activity concentrations, of which five samples also measured elevated alpha activity. All the groundwater samples that exhibited elevated radioactivity were of the HCO_3-Na type and were genetically associated with granitic rocks. Their TDS levels and CO_2 gas concentrations were also elevated.

Key words: radioactivity, activity concentration, ^{40}K, groundwater, Serbia.

Апстракт. Концентрације радионуклида ^{40}K, ^{228}Ra, ^{226}Ra, ^{238}U и ^{232}Th, као и укупна алфа и бета активност су анализирани у више од 100 узорака подземних вода са територије Србије. Укупна алфа активност достиже максимално 1,33 Bq/L са средњом вредношћу 0,12 Bq/L. Укупна бета активност има максималну вредност од 5,43 Bq/L, са средњом вредношћу од 0.68 Bq/L. Највише концентрације активности има изотоп калијума, ^{40}K који има и највећи допринос природној бета активности. Од испитаног броја подземних вода, 28 узорака је показало повишене концентрације бета активности, а пет узорака има повишене вредности алфа активности. Све испитиване воде које се одликују повишеном радиоактивношћу су HCO_3-Na типа, и генетски су највероватније везане за гранитне стенске масе. Такође, у њима је повишен садржај CO_2 као и растворених минералних материја.

Кључне речи: радиоактивност, активне концентрације, ^{40}K, подземна вода, Србија.

Introduction

The discovery of radioactivity and its impacts was a turning point in the evolution of geological sciences and largely affected the development of geochemistry, including isotope geochemistry, and geochronology that plays an important role in terrestrial geology. A special discipline is environmental geology that studies the impact of human activity on the environment, as well as that of natural radioisotopes (Omaljev & Antonović 1996). Radioactivity is an important parameter of geophysical measurements in gamma prospecting, logging of natural gamma radioactivity (gamma logging) and spectral gamma logging.

Prior to the discovery of radioactivity, it was believed that the main cause and source of heat flow within the Earth was cooling of its previously heated body (Marović 2005). Apart from the heat content of the Earth immediately after formation, the radiogenic decay of the unstable isotopes of uranium (^{238}U, ^{235}U), thorium (^{232}Th), and potassium (^{40}K) provides the largest internal source of heat (Clauser 2011; Hazen *et al.* 2009). The release of heat was likely more intense in the Earth's distant past than today, because the amount of radioactive elements constantly decreases as a result of decay (Marović 2005). Uranium, thorium and ^{40}K, are no longer trapped in the planet's core, they migrated to the Earth's surface in the early stages because of the crystalline and chemical properties of the compounds into which they were incorporated (Antonović 1989).

Origin of radioactivity in nature

The main α emitting radionuclides in the natural decay series are ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Po, ^{232}Th and ^{228}Th (Turhan 2013). Positively charged parti-

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cles, helium cores or ions comprised of two protons and two neutrons, make up gross alpha activity. Gross beta activity is comprised of negatively charged particles (electrons) or, rarely, positively charged particles (positrons). Nearly all radionuclides are beta emitters, except for some of the heaviest cores, while the major β emitting radionuclides are ^{210}Pb and ^{228}Ra as well as ^{40}K (TURHAN 2013). Beta emitters are generally also gamma emitters, with the exception of some pure beta (β^-) emitters such as ^{14}C, ^{45}Ca, ^{63}Ni, ^{90}Sr, ^{106}Rh and ^{147}Pm (ICRP 1991).

Environmental radiation originates from a number of naturally occurring and man-made sources. Radioactive materials occur naturally anywhere in the environment (for example uranium, thorium and potassium) (WHO 1993).

Elements 84 through 92 on the periodic table have no stable isotopes at all and can be grouped into one of three radioactive series or families. These are the uranium-radium, uranium-actinium and thorium series. Natural radioactive series are a result of decay of three radioisotopes: ^{235}U, ^{238}U and ^{232}Th, ending with isotopes ^{206}Pb, ^{208}Pb, ^{207}Pb and ^{209}Bi (USGS, 1998). The radiation generated though the decay of these naturally-occurring radioactive isotopes is the natural radioactivity found in our environment (ICRP 1991).

The radioactivity in groundwater comes mainly from radionuclides of the natural decay chains ^{238}U and ^{232}Th, and ^{40}K in soil and bedrock. Some radionuclides can migrate easily in water, depending on the mineralogical and geochemical composition of the soil and rock, redox conditions and the residence time of ground water in the soil and bedrock, as result of the reaction of the ground water with soil and bedrock (VESTERBACKA 2007).

The territory of Serbia is rich in thermal and mineral waters. Because of the volcanic and plutonic activity in the geological past, most of the mineral or thermal water originates from these rocks (PROTIĆ 1995). Groundwater occurrences mark different regional geological-structural features and the largest number of mineral groundwater is related to the granite intrusions and volcanic rocks (MARINKOVIĆ *et al.* 2013). A major portion of uranium and thorium in igneous rocks is concentrated in accessory minerals such as zircon, sphene and apatite. Other highly radioactive minerals (e.g. monazite, alanite, pyrochlore, xenotime and thorite) are found only occasionally. In general, uranium and thorium concentrations in igneous rocks increase with increasing rock acidity. The major radioactive minerals are shown in Table 1.

Table 1. Minerals and rocks featuring radioactive elements (ANTONOVIĆ 1989).

Radioactive element	Radioactive mineral	Rock/process
Potassium	Orthoclase and microcline feldspars ($KAlSi_3O_8$)	Main ingredient of acidic rocks and pegmatites
	Muscovite ($Na_2KAl(SiO_4)_3$)	As above
	Alunite ($KAl_3(SO_4)_2(OH)_6$)	Alterations in acidic volcanites
	Sylvite, carnallite (KCl, $MgCl_2 \cdot 6H_2O$)	Deposits in salt sediments
Uranium	Uraninite (oxide of U, Pb, Ra+Th, rare earth elements)	Granites, pegmatites with Ag, Pb and Cu veins
	Carnotite ($K_2O \cdot 2UO_3 \cdot V_2O_5 \cdot 2H_2O$)	Sandstones
	Gummite (alteration of uraninite)	Together with uraninite
Thorium	Monazite (Th_2O + rare earth phosphates)	Granite, pegmatite, gneiss
	Thorianite ($(Th, U)O_2$)	Granite, pegmatite
	Thorite uranothorite ($ThSiO_4$+U)	Granite, pegmatite

Numerous radioactive elements and isotopes occur in natural waters: uranium, radium, radon (descendants of uranium and thorium), while thorium is virtually absent because of its low mobility in geochemical systems where groundwater reservoirs develop (DANGIĆ 1995).

Methods

Sampling

During the course of the study: Radioactivity of Groundwater in the Republic of Serbia (PAPIĆ *et al.* 2008–2011), gross alpha and beta activities and the radionuclides (^{40}K, ^{228}Ra, ^{238}U, ^{226}Ra and ^{232}Th) in

more than 100 groundwater samples were analyzed and represented in this paper. Sampling was conducted applying standard closed-source methods and suitable polyethylene containers were filled with 10–15 L of water.

Determination of gross alpha/beta activity concentrations

The gross alpha and beta activities of groundwater samples and gamma spectrometry measurements of natural radionuclides (^{238}U, ^{228}Ra, ^{226}Ra, ^{40}K, ^{232}Th) have been carried out in the Institute of Occupational and Radiological Health "Dr Dragomir Karajović". The gross alpha/beta analyses were performed according to a routine procedure outlined in ISO9696 and ISO9697 (ISO 9696, 1992; ISO 9697, 1992).

The gross alpha/beta activity determination method was based on the evaporation of 3 L of the water sample under UV lamps and calcining at 550 °C to a constant mass. The mineral residue was used to prepare a thin-film test sample. Suitable-geometry measurements were made on a low level proportional counter Thermo-Eberline FHT 770T, featuring a 21% efficiency for alpha radiation and 33% for beta radiation. The instrument was calibrated using standard sources. The sample measurement time was 3600 s. The measurement results were used to determine gross alpha and beta activities of the water samples in Bq/L.

Table 2. Maximum permissible concentration (MPC), average, minimum and maximum activity concentrations of radionuclides, standard deviation and median values and gross alpha and beta activities in groundwater samples collected across Serbia.

Parameter (Bq/L)	No. of samples	MPC (Bq/L)	Average	Min	Max	Std.Deviation	Median
α	125	**0.5**	0.12	0.001	1.33	0.21	0.04
β	125	**1**	0.68	0.018	5.43	0.93	0.26
^{40}K	116	**-**	0.56	0.012	2.6	0.63	0.24
^{228}Ra	116	**0.2**	0.10	0.006	0.76	0.16	0.05
^{238}U	116	**3**	0.15	0.010	0.80	0.12	0.12
^{226}Ra	116	**0.49**	0.16	0.005	2.56	0.36	0.04
^{232}Th	47	**0.59**	0.08	0.006	0.79	0.16	0.04

Determination of radionuclide activity concentrations

The method for the determination of gamma-emitter radionuclide activity was based on the evaporation of 8–10 L of the water sample to a volume of 200 mL and quantitative transfers to polyethylene vessels. The vessels were closed and left to stand for about 40 days, to prevent radon emanation and establish a radioactive balance between members of the natural radioactive series of ^{238}U. The sample was then measured on a gamma spectrometer with a HP Ge detector, whose relative efficiency was 23%. The detector was calibrated using standard radioactive reference material, MIX-OMH-F. The duration of sample measurements was 1.3 days, depending on the concentrations present. The gamma radiation spectrum was analyzed to determine the concentrations of specific radionuclides (^{40}K, ^{228}Ra, ^{226}Ra,^{238}U and ^{232}Th) in Bq/L.

Chemical analyses were performed at the Hydrochemistry Lab of the University of Belgrade Faculty of Mining and Geology to define hydrogeochemical conditions and determine groundwater types. The following parameters were analyzed: sodium, potassium, calcium, magnesium, chlorides, hydrocarbonates, carbonates, sulfates, TDS, hardness, pH, specific conductivity and CO_2.

The results were statistically processed using IBM software SPSS 17.0 (Inc SPSS 2009) and graphically interpreted by ESRI ArcGIS 9.3 (ESRI 2012).

Results and discussion

Basic statistical processing, including active concentration ranges, median and standard deviation (Table 2), was conducted on the basis of 125 analyses of groundwater samples for gross alpha and beta activities and a certain number of active concentrations of radionuclides ^{40}K, ^{228}Ra, ^{238}U, ^{226}Ra and ^{232}Th.

The World Health Organization, following the recommendations of the International Commission on Radiological Protection (ICRP), has studied the radiological aspects of drinking water quality and recommended reference values for α-unstable radionuclides in drinking water of 0.5 Bq/L, and β-unstable radionuclides of 1 Bq/L (ICRP 1991; WHO 1993; Official Gazette of RS 2011). When the average and median values were compared to regulated MPC (maximum permissible concentration) levels, the groundwater samples did not exhibit elevated radioactivity. However, when the maximum values of the individual parameters were evaluated, gross alpha and beta activities and the concentrations of ^{228}Ra and ^{226}Ra were found to exceed permissible concentrations. Histograms of gross alpha and beta activities and active concentrations of the tested radionuclides were produced to determine the cumulative distributions of the analyzed parameters (Fig. 1).

The histograms of gross alpha and beta activities and active concentrations of unstable radionuclides showed that all distributions exhibited positive skewness. Only five samples measured gross alpha activity in excess of the MPC of 0.5 Bq/L, while 28 samples

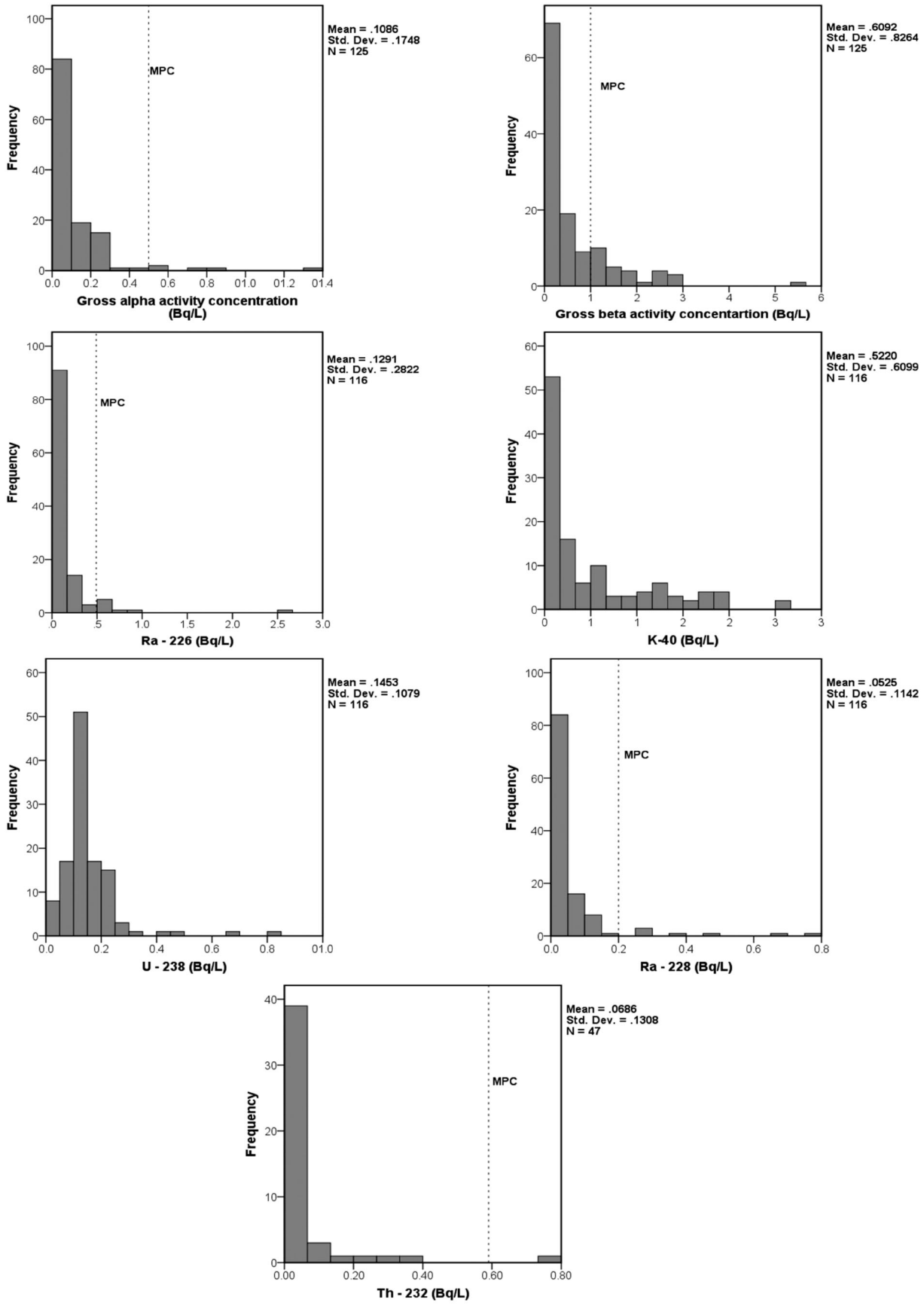

Fig. 1. Histograms of gross alpha and beta activities and active concentrations of radionuclides ^{40}K, ^{226}Ra, ^{228}Ra, ^{232}Th and ^{238}U.

were above the beta activity threshold of 1Bq/L. Radionuclides ^{226}Ra, ^{228}Ra and ^{232}Th exceeded MPC levels only in a few isolated cases, deemed to be extremes or outliers for statistical interpretation purposes and therefore disregarded. There is no regulated MPC level for ^{40}K in Serbian regulations;the average value of the samples is 0.56 Bq/L. The distribution of ^{40}K was found to be similar to that of gross beta activity and this radionuclide was the greatest contributor to gross beta activity, corroborated by high coefficient of correlation ($R^2 = 0.844$) between gross beta activity and the potassium isotope ^{40}K. The concentration of ^{40}K was found to be consistent with the geochemistry of potassium, which is one of the main elements of magma (MITTLEFELDHT 1999).

The most frequently encountered radioactive potassium minerals are orthoclase and microcline feldspars ($KAlSi_3O_8$) and muscovite ($Na_2KAl(SiO_4)_3$), which are the main minerals of acidic igneous rocks and pegmatites. Alunite ($KAl_3(SO_4)_2(OH)_6$) occurs though alteration in acidic volcanites, while sylvite and carnallite (KCl, $MgCl_2 \cdot 6H_2O$) are deposits found in salt sediments (ANTONOVIĆ 1989). ^{40}K also exhibited a high coefficient of correlation ($R^2 = 0.43$) with gross alpha activity but this result was not unexpected given that the TDS levels of the tested groundwater samples were up to 6650 mg/L. These groundwaters were formed in deep hydrogeological structures. Figure 2 shows that gross active concentrations of alpha/beta emissions increased with increasing TDS.

Both correlations between α/β activity and the concentrations of TDS (Total dissolved solids) with a coefficient of R^2=0.36 for gross alpha activity (Fig. 2A) and R^2=0.42 for gross beta activity (Fig. 2B).

The samples were divided into two groups to better understand the hydrogeochemical conditions in which the groundwaters that exhibited elevated radioactivity were formed. One group was comprised of 28 groundwater samples whose active alpha or beta concentrations exceeded MPC, and the other group was made up of 97 samples that measured below MPC. Select chemical composition parameters are presented in box-plots to show the general differences between these two groups (Fig. 3).

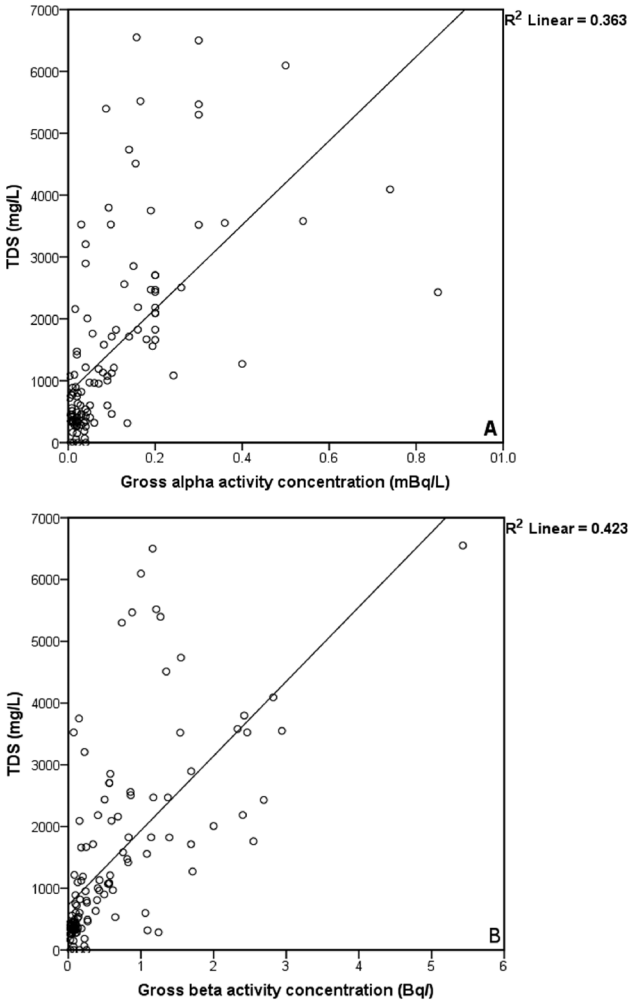

Fig. 2. **A,** Correlation between gross alpha activity and dissolved minerals. **B,** Correlation between gross beta activity and dissolved minerals.

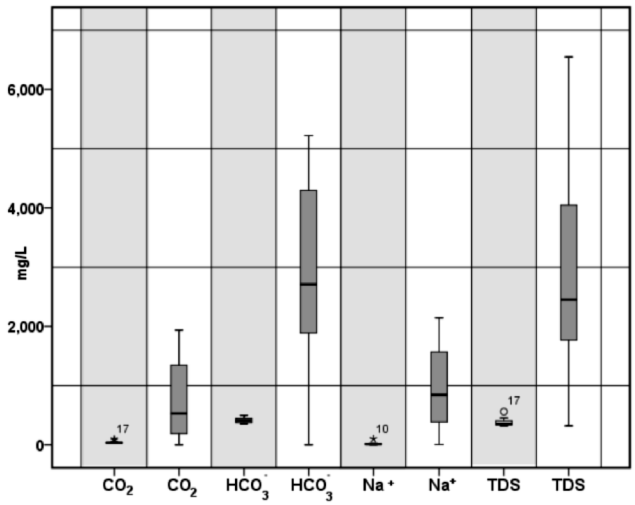

Fig. 3. Box plots of CO_2, HCO_3^- and Na^+ concentrations and TDS for two groups of groundwater samples (the group of 97 samples is shaded).

All the tested samples that exhibited elevated beta activity concentrations belonged to the Na-HCO_3 type of groundwater, suggesting that the groundwater traced to an aquifer in granitoid rocks with elevated concentrations of dissolved solids and CO_2. The samples whose radioactivity was elevated measured CO_2 concentrations up to 1510 mg/L. The water samples whose mineral content (TDS) was 500 mg/L, typical of groundwater that did not show elevated radioactivity, were dominated by Ca^{2+} and Mg^{2+} ions, while with increasing TDS these ions were replaced with the Na^+ ion.

Gas CO_2 in groundwater may be derived from a variety of sources, including metamorphic devolatilisation, magmatic degassing, oxidation of organic mat-

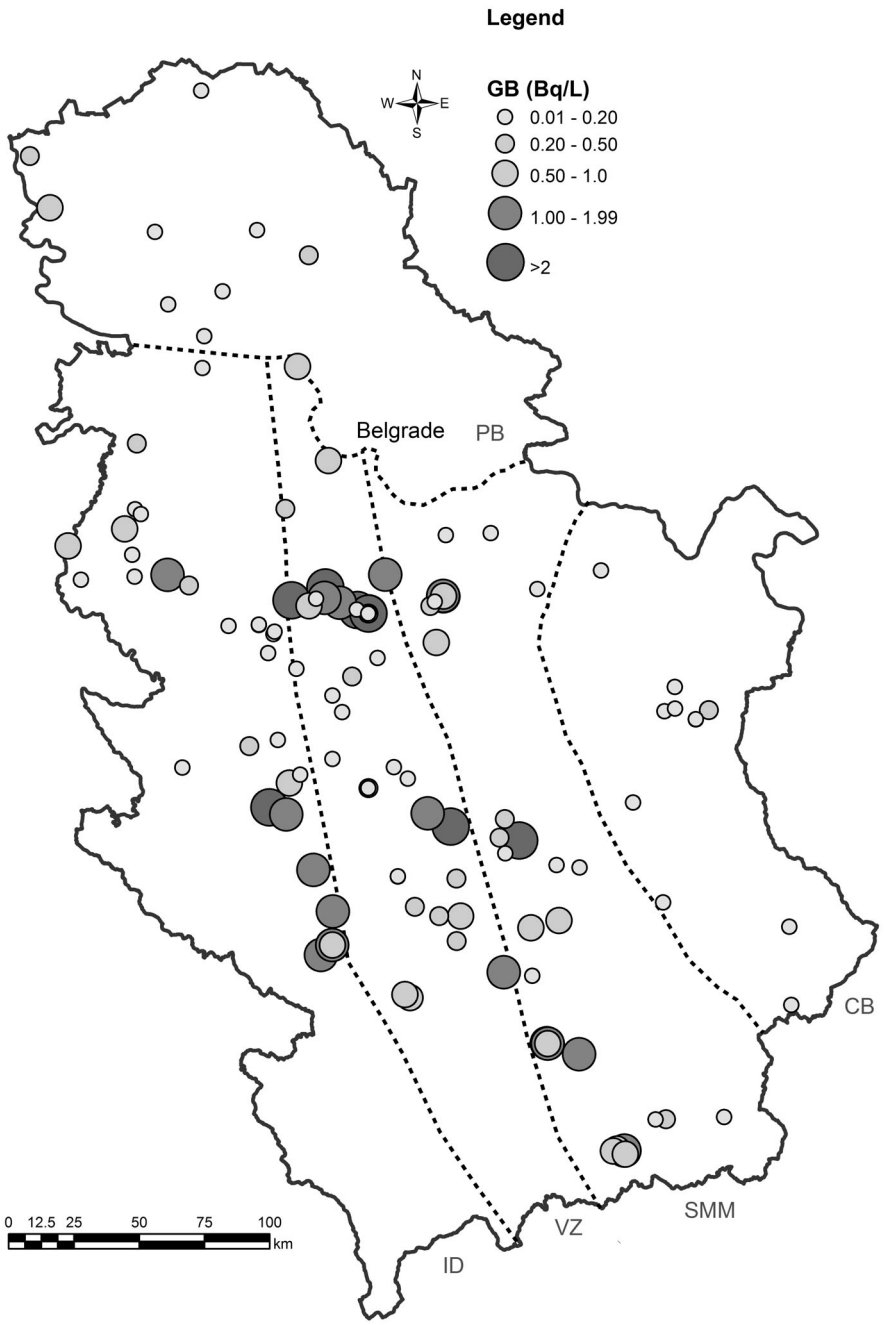

Fig. 4. Active concentrations of gross beta activity in groundwater relative to well-known geological/tectonic units. **VZ,** Vardar Zone; **SMM,** Serbo-Macedonian Massif; **ID,** Inner Dinaride; **CB,** Carpatho-Balkanides; **PB,** Pannonian Basin.

ter and interaction of water with sedimentary carbonates (CARTWRIGHT *et al*. 2001). Research conducted to date has shown that the most important radioactivity anomalies are found in calc-alkaline igneous rocks (JELENKOVIĆ 1991), which are rich in alkaline oxides and metals and make up most of the continental crust. Past studies of carbonated groundwater have shown that groundwater with elevated CO_2 concentrations tends to occur in areas of large tectonic faults, in zones of Tertiary and Quaternary volcanism and within regional metamorphism. These formation must have been related to the part of the lithosphere in which the CO_2 gas was generated. (STEVENS *et al.* 2001; MARINKOVIĆ *et al.* 2012, 2013). Spatial distribution of gross beta activity is apparent in Figure 4, and shows that most of the groundwater samples that exhibit elevated beta activity are located in the Vardar Zone or at the interface between the Vardar Zone and the Serbo-Macedonian Massif (SMM) and the Inner Dinarides (ID). This studys include 11 occurrences of groundwater from the area of the Pannonian Basin (PB) and all samples have beta activity concentrations less than 1 Bq/L. Also the area of the Carpatho-Balkanides (CB) covered by the 10 groundwater samples is characterized by low values of total beta activity.

Previous research has shown that certain deposits of radioactive elements can be spatially associated with volcanic complexes, being favorable environments for the creation of such deposits, but that the deposits themselves are linked with igneous reservoirs. It has been established that these deposits, and consequently the places where groundwater with elevated concentrations of radioactive elements is found, can occur in areas associated with large regional geotectonic zones, usually following their direction (RISTIĆ 1969). The main CO_2 generators are located within the Vardar Zone that is characterized by a highly complex geological/tectonic makeup and Tertiary magmatism and associated with the occurrence of carbonated groundwater (MARINKOVIĆ *et al*. 2012). This groundwater also exhibits naturally elevated radioactivity.

Conclusion

In this paper the distribution of natural radioactivity of groundwater from the territory of Serbia is presented, which has significant regional importance.

Determination of chemical composition and the concentration of radionuclides in groundwater may contribute in various fields within geology and hydrogeology. Gross alpha and beta activities of 125 groundwater samples collected in Serbia were determined; 28 samples featured elevated beta activity, of which 5 samples also measured elevated alpha activity. The origin of the radioactivity of these groundwater samples is natural, tracing to accessory minerals in igneous rocks. The major β-emitting radionuclide in the analyzed samples was ^{40}K, which exhibited a very high coefficient of correlation with gross beta activity. This radionuclide adheres to the geochemistry of potassium, which is one of the main magma constituents. The samples that exhibited elevated radioactivity were of the HCO_3-Na type of groundwater whose TDS exceeded 500 mg/L and CO_2 concentrations measured 33–1510 mg/L. Most groundwater occurrences that feature elevated radioactivity appear to be found in the Vardar Zone or at the interface between the Vardar Zone and adjacent zones.

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Резиме

Природна радиоактивност подземних вода у Србији

Радиоактивни елементи, потомци урана и торијума, се у великој мери јављају у природним водама. Велики део ових елемената се налази у магматским стенама и концентрисан је у пратећим (акцесорним) минералима као што су циркон, сфен и апатит. Генерално, садржај урана и торијума у магматским стенама расте са киселошћу стена. Основни радионуклиди који емитују α зрачење су ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Po, ^{232}Th и ^{228}Th, док су најважнији β радиоануклиди ^{210}Pb, ^{228}Ra као и ^{40}K.

У циљу сагледања радиоактивних особина подземних вода са територије Србије анализирано је више од 100 појава подземних вода и одређиване су концентрације радионуклида ^{40}K, ^{228}Ra, ^{226}Ra, ^{238}U и ^{232}Th, као и укупна алфа и бета активност подземних вода. Међутим, да би се разматрале радиоактивне особине подземних вода, било је потребно проучити и основни хемијски састав вода. У Лабораторији за хидрохемију, Рударско-геолошког факултета урађене су хемијске анализе ради дефинисања хидрогеохемијских услова и утврђивања типова подземних вода. Како би се утврдила веза за типом подземних вода у којој се јављају повишене концентрације радионуклида у води, одређивани су следећи хидрохемијски параметри: натријум, калијум, калцијум, магнезијум, хлориди, хидрокарбонати, сулфати, укупна минерализација, тврдоћа, pH вредност и садржај гаса CO_2. Због великог броја појава прво је приступљено утврђивању основних статистичких параметара радионуклида и алфа и бета активности (максималне, минималне концентрације, средње вредности и стандардне девијације). Од испитаног броја подземних вода, 28 узорака, је показало повишене концентрације бета активности, од чега 5 узорака има повишене вредности алфа активности. Средња вредност укупне алфа концентрације износи 0,12 Bq/L, док је за бету 0,68 Bq/L. Очигледно је да укупна бета активност представља највећим делом узрок повишене радиоактивности, а ^{40}K је радионуклид који у највећој мери доприноси укупној бета активности. Средња вредност ^{40}K износи 0,56 Bq/L.

Како би се дефинисао тип вода који се карактерише повишеном природном радиоактивношћу, сви испитивани узорци подземних вода су подељени у две групе, тако да једну групу чине воде чија укупна алфа активност прелази 0,5 Bq/L, односно укупна бета 1 Bq/L. Свих 28 појава које прелазе прописане максималне дозвољене вредности укупне алфа и бета активности припадају Na-HCO_3 типу воде, што указује на могућност порекла вода из аквифера који се налази у гранитоидним стенама. Уједно, ове воде се одликују повишеним садржајем растворених минералних материја и гаса CO_2. Угљен–диоксид се у узорцима са повишеном радиоактивношћу налази у опсегу 33–1510 mg/L. Подземне воде чија је минерализација мања од 500 mg/L имају доминантне јоне Ca^{2+} и Mg^{2+} (што је карактеристично за узорке који нису имали повишену радиоактивност), док се са повећањем минерализације ови јони смењују са Na^{+} јонима. Самим тим се може закључити да се са повећањем растворених минералних материја у води повећава и радиоактивност подземних вода, што указује на то да се радиоактивност подземних вода везује за геолошке формације у којима су формирани аквифери.

Претходним истраживањима констатовано је да се најважније аномалије радиоактивности везују за калко-алкалне магматске стене, које чине највећи део континенталне коре, односно за области великих тектонских раседа, у зони терцијарног и квартарног вулканизма и у оквиру регионалног метаморфизма. Овим радом је представљена просторна дистрибуција укупне бета активности на територији Србије, где се уочава да је највећи број подземних вода са повишеном бета активношћу смештен у Вардарској зони или на контакту Вардарске зоне са Српско-македонском масом и Унутрашњим Динаридима.