

## Distribution of magnesium in groundwater of Serbia

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**Abstract.** Magnesium is chemical element commonly found in the environment and the main constituent of many types of minerals and rocks. This element is also essential to man. Owing to its abundance in nature, magnesium is present in all water resources and generally occur as the dominant cation, with calcium, in those that feature low TDS levels, whose origin is associated with large formations of sedimentary rocks (limestones, dolomites), and to a lesser extent with the degradation of silicate minerals that contain Mg. Magnesium concentrations in groundwater of Serbia vary over a wide range and their distribution is not uniform, but certain laws of nature do apply. The variation in the concentrations of this ion depends on the considered hydrogeological province, while within a single province it is a consequence of Serbia's highly complex geology. The best examples are the Carpatho-Balkanides, with predominant karstified rock formations, and the Vardar Zone where ophiolites prevail but the makeup is much more complex than that of the Carpatho-Balkanides.

**Key words:** magnesium, groundwater, distribution, Mg/Ca ratio, Serbia.

**Апстракт.** Магнезијум је земноалкални метал, веома заступљен у животној средини и главни конституент многих типова минерала и стена, а уједно есенцијалан за човека. Захваљујући великој распрострањености у природи, присутан је у свим водама и обично се са калцијумом, јавља као доминантни катјон у маломинерализованим водама, чије се порекло везује за огромне масе седиментних стена (кречњаци, доломити) и мањим делом за процесе распадања силикатних минерала који садрже магнезијум. Садржај магнезијума у подземним водама Србије варира у веома широком опсегу, дистрибуција је неравномерна, али се могу уочити одређене закономерности. Варирање концентрација магнезијума у зависности од посматраног рејона, и у оквиру једног рејона је последица веома сложене геолошке грађе Србије. Ово најбоље потврђују примери рејона Карпато-балканела, са доминантним распрострањењем карстификованих стенских маса и рејон Вардарске зоне са доминантним распространењем оphiолита, али и знатно сложенијом грађом од Карпато-балканела.

**Кључне речи:** магнезијум, подземне воде, дистрибуција, однос rMg/rCa, Србија.

## Introduction

Magnesium is strongly lithophile, the eight most common element in the upper lithosphere. Unlike the alkali metals, it is able to build both simple and complex inorganic compounds which are stable under the conditions met in nature (RANKAMA & SAHAMA 1950). Magnesium is a significant component of most rock systems and a major constituent of many rock forming minerals (MITTLEFEHLDT 1999; JOVIĆ & JOVANOVIĆ 2004). The chief magnesium-containing minerals are the olivine series, garnets, the pyroxene, amphibole and mica groups, chrysotile, sepiolite, talc, serpentine, the chlorite group, and magnesium-bearing clay minerals. During chemical weathering magnesium is released. Important amounts of magnesium are con-

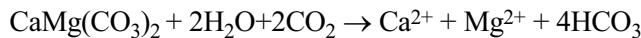
tained in dolomite and magnesite, and mixtures of these in limestone. Therefore greater geochemical abundance of Mg occur in Mg-rich aquifers, such as olivine-basalts, serpentines, and dolomite rocks; however the absolute magnesium contents in these cases are also low. Minerals with exchange capability adsorb Mg only slightly more firmly than Ca, so that low Mg contents can occasionally be attributed to cation exchange (DAVIS & DEWIEST 1966).

Estimates of Mg concentrations in the lithosphere vary from 132 to 158 mg/g; the highest Mg concentrations tend to be found in ultramafic rocks (HITCHON *et al.* 1999). In unpolluted shallow groundwater, magnesium concentrations range from 0.1–1.2 to about 50 mg/L (COX 1995).

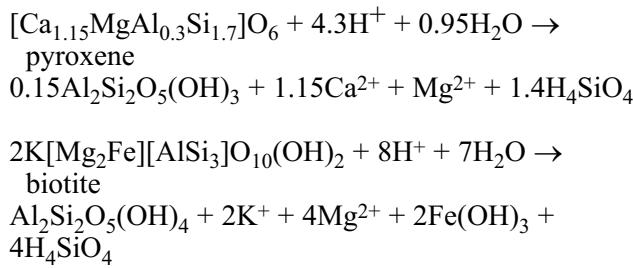
In a sedimentation environment, Mg largely occurs

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in association with the carbonate ion, predominantly as dolomite  $\text{CaMg}(\text{CO}_3)_2$ . In the hydrosphere,  $\text{Mg}^{2+}$  enters by the weathering process, mainly through decomposition of dark ferromagnesian minerals, chlorite, Mg-calcite and dolomite (HITCHON *et al.* 1999). Dolomite dissolution and entering Mg in hydrosphere on this way is described by reaction (APPELO & POSTMA 1996):



The second important way of groundwater magnesium enrichment is the silicate weathering, particularly of ultramafic rocks made up largely of magnesium silicates, which react rapidly with water (BARNES *et al.* 1978). The most common weathering reaction on earth is the process of hydrolysis, producing new minerals as well as relocation of Mg ions into solution. Minerals which are undergone on this process are relative insoluble, and the final products of this process could be orthosilicic acid and clay minerals. Reactions of pyroxene and biotite decay are shown below (APPELO & POSTMA 1996).



Magnesium plays a multiple roles in the human body: it acts directly on the neuromuscular plate, is essential for normal vitamin C and vitamin B<sub>1</sub> activity, takes part in enzymatic processes leading to energy production, reduces coagulation levels, protects the inner walls of blood vessels from fibrosis, and catalyzes the utilization of fats, proteins and carbohydrates (TEOFILOVIĆ *et al.* 1999; TOUYZ & SONTIA 2009; JAHHEN-DECENT & KETTELER 2012).

Despite the many positive effects and multiple roles in the human body, very large doses of magnesium could have some negative effects. At large oral doses magnesium may cause vomiting and diarrhea, but there are no known cases of magnesium poisoning (MAS-SRY & SEELIG 1977; SINGH 2010). Too much magnesium does not pose a health risk in healthy individuals because the kidneys eliminate excess amounts in urine (MUSSO 2009). Nevertheless, national drinking water standards have limited the concentration of magnesium in drinking water at 50 mg/L (OFFICIAL GAZETTE OF THE SERBIA AND MONTENEGRO 53/2005).

The geology of Serbia is highly complex and not conducive to generalized studies and assessments. To facilitate insight, hydrogeological provinces have been identified. This hydrogeological zoning is based on geotectonic units, with respect to historical and geological processes, structural-geological conditions, petrological characteristics, geomorphological, physical, geographical, hydrological, hydrometeorological, hydrogeological and other conditions of the environment. In broadly general terms, the provinces are (FILIPović *et al.* 2005): the Dacian Basin, the Carpatho-Balkanides, the Serbian Crystalline Core, the Vardar Zone, the Inner Dinarides and the Pannonian Basin (Fig. 1).

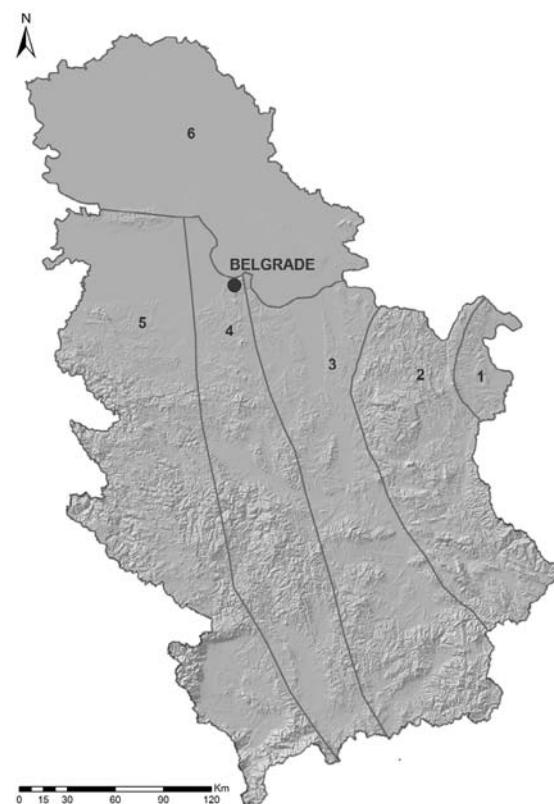


Fig. 1. Hydrogeological provinces of Serbia. Legend: 1, Dacian Basin; 2, Carpatho-Balkanides; 3, Serbian Crystalline Core; 4, Vardar Zone; 5, Inner Dinarides; 6, Pannonian Basin.

The Carpatho-Balkanides are mostly composed of Mesozoic limestones and dolomites which are thick more than 1000 m. Another province, the Serbian Crystalline Core occupies the central part of the territory and it can be divided into two parts. In the north part of Serbian Crystalline Core the most dominant are Neogene and Quaternary sediments in the river valleys, while the south part of Serbian Crystalline Core is composed of very thick Proterozoic metamorphic rocks: gneiss, micaceous shale, various types of schist, marble, quartzite, but also of igneous rocks (intrusive-granitic and volcanic rocks of Tertiary age). The Vardar Zone separates the Serbian Crystalline

Core and the Inner Dinarides. It is characterized by a complex structure consisting of medium grade metamorphosed schist, recrystallized limestones and marbles. In the Inner Dinarides, there is a significant occurrence of Triassic limestones and dolomites, followed by the Jurassic diabase-chert formation (ophiolitic belt) with subordinate limestones in the overlying parts, and the Cretaceous formations with predominantly flysch. The Pannonian Basin, or its south-eastern part in Serbia, consists of Palaeogene, Neogene and Quaternary sediments with a total maximum thickness of about 4000 m (PETROVIĆ *et al.* 2012; FILIPOVIĆ *et al.* 2005).

Considering that the Dacian basin has no natural occurrence of groundwater on the surface, but only the presence of highly mineralized chloride-sodium groundwater (brine) with a depth of over 1000, the groundwater occurrences of this zone are not taken into further consideration (FILIPOVIĆ 2003).

## Approach and method

The data used in this research were derived from investigations conducted from 2008 to 2012. Groundwater was sampled at 253 locations across Serbia, including groundwater resources featuring low and high total dissolved solids (TDS) levels. The sampling network was designed to evenly cover the entire territory of Serbia and address groundwater occurrences in different rocks (igneous, metamorphic and sedimentary), and consequently different types of aquifers. The sampling points included springs, boreholes and wells. Sampling was conducted in accordance with the Drinking Water Sampling and Laboratory Analysis Rulebook (OFFICIAL GAZETTE OF THE SFRY 33/87). All groundwater samples were tested to determine the main physicochemical parameters (temperature, pH, electrical conductivity) and the basic chemical composition. The analyses were conducted at the Hydrochemistry Lab of the University of Belgrade Faculty of Mining and Geology, as well as at the Public Health Institute of Belgrade. Magnesium concentrations were determined by the ICP-OES method.

Chemical analyses of groundwater samples were statistically processed to assess and interpret hydrochemical data and to generate hydrochemical maps of magnesium distribution in groundwater of Serbia. The data were statistically processed and graphically interpreted using statistical software IBM SPSS v.19. The hydrochemical maps of magnesium distribution in groundwater of Serbia scale 1:500 000 were generated using ESRI ArcGIS 10.0 software.

## Results and discussion

### General groundwater quality

Serbia's highly complex geology has resulted in groundwater resources featuring different types, tem-

peratures and TDS levels. The dominant anion in the analyzed groundwater samples is the hydrocarbonate ion. Apart from several occurrences of sulfate, chloride, hydrocarbonate-sulfate and hydrocarbonate-chloride types, more than 90 % are found to be of the hydrocarbonate type of groundwater. Of all the analyzed samples, three belong to the sulfate type and two to the chloride group with a chloride share of 97 %equ. The latter two occurrences featured high TDS levels, in excess of 6 g/L. Based on their cation composition, the samples predominantly reflect Ca, Na and composite (Ca-Na, Ca-Mg, Ca-Mg-Na) types of groundwater. Four samples are of the Mg type, with a magnesium share in excess of 75 %equ (Fig. 2).

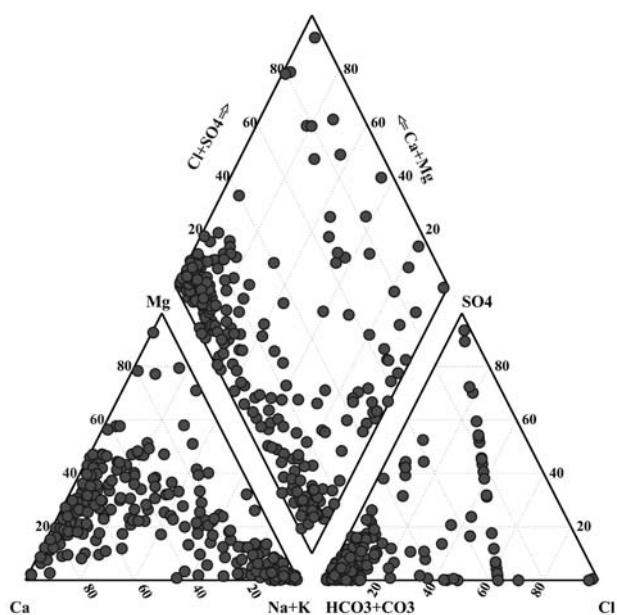


Fig. 2. Piper diagram of the chemical composition of groundwater samples.

With regard to total dissolved solids (TDS), the analyzed samples exhibited considerable diversity: from low levels (only 29 mg/L) to very high levels (in excess of 8 g/L).

### Magnesium in groundwater of Serbia

Magnesium concentrations in Serbia's groundwater resources vary considerably, from 0.07 mg/L to 324 mg/L. The average is 32.10 mg/L. Magnesium concentrations in groundwater depend on the geology and tectonics of the ground in which the groundwater is formed, as well as the type of groundwater and the TDS level, given that the concentration of this ion is higher in high-TDS than in low-TDS groundwater, such that this groundwater is not of a pure Mg type.

In most of the samples (121 or 48 %), Mg concentrations are only up to 20 mg/L, while 36 % or 92 samples

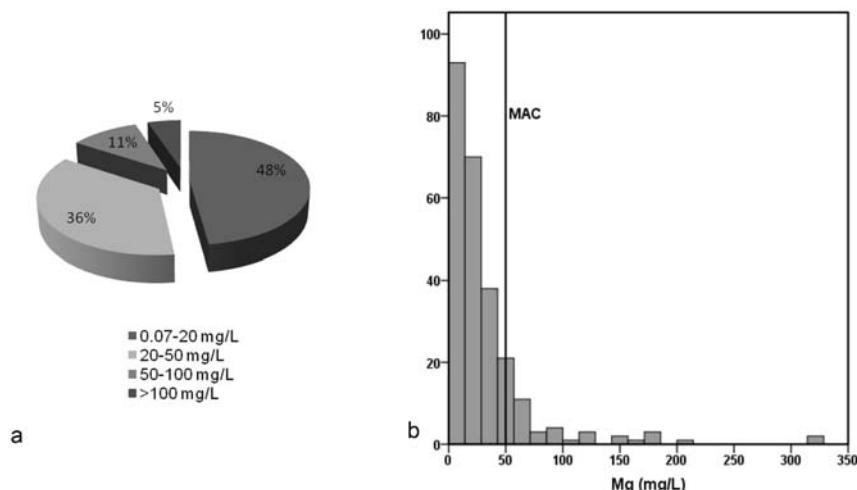


Fig. 3. Plot of relative frequency of Mg (a) and histogram of Mg concentrations in Serbia's groundwater resources (b).

contain 20 to 50 mg/L (Fig. 3a). This means that 84 % of the groundwater samples collected across Serbia exhibited Mg concentrations up to 50 mg/L, which is the maximum allowable concentration (MAC) according to national drinking water standards (OFFICIAL GAZETTE OF THE SERBIA AND MONTENEGRO 53/2005).

As the diagram (Fig. 3b) shows, of the 253 samples, 40 samples (or 16 %) have Mg concentrations in

Table 1. Magnesium concentrations in different hydrogeological provinces (mg/L).

Province	Number of samples	min	avr	max
Carpatho-Balkanides (2)	27	0.08	14.40	29.00
Serbian Crystalline Core (3)	26	0.07	40.37	183.00
Vardar Zone (4)	55	0.73	46.33	324.00
Inner Dinarides (5)	67	1.08	22.91	86.00
Pannonian Basin (6)	78	5.60	26.45	65.30

excess of drinking water standards. In view of the total number of samples analyzed and the fact that the MAC for magnesium is considerably lower than for other main cations, such a proportion was expected.

Mg concentrations above 50 mg/L, are found in high-TDS groundwater with TDS more than 1 g/L, where some samples has TDS up to 6 g/L. Such groundwater are generally traced to schists.

In several samples of low-TDS groundwater, Mg concentrations measure 50–60 mg/L; such groundwater is largely genetically associated with rocks like dolomite and dolomitic limestone, as well as interfaces of these rocks with Neogene sediments, flysch, fractured and degraded sandstones and marls, or rocks whose composition includes magnesium-rich minerals. Occurrences of low-TDS groundwater, where Mg concentrations are in the 60–70 mg/L range, were associated with serpentinites, fractured harzburgites and

dolomite/serpentinite contacts. This occurrences are usually groundwaters of Mg type.

The distribution of magnesium in groundwater of Serbia is not uniform (Fig. 4.), but regularities resulting from the geological makeup are much more apparent.

Table 1 shows the smallest variations but also the lowest Mg concentrations in the Carpatho-Balkanide Province, where the average Mg concentration is found to be 14.40 mg/L. This province is dominated by limestone and dolomitic karstified rock formations (STEVANOVIC 1991) as corroborated by Mg to Ca

ratio, hence, maximum Mg concentration of only 29 mg/L is usual.

The Inner Dinarides of western Serbia exhibited an average Mg concentration in groundwater of 21.91 mg/L; the concentrations of this ion are generally below the MAC, even though there are several exceptions whose concentrations are moderately higher (86 mg/L). The occurrences that give Mg concentrations up to the average level (about 22 mg/L) are generally low-TDS groundwater resources tracing to the limestone formations of this province, with a slight influence of diabase-chert rocks or Neogene sediments. Mg concentrations from the average level to 50 mg/L are recorded in samples collected from groundwater occurrences in limestones under consider-

able influence from serpentinites, harzburgites and similar rocks that make up the Dinaride ophiolite zone. This province feature a distinct occurrence of groundwater of the Mg type associated with pure serpentinites and harzburgites. This is low-TDS groundwater (350 mg/L), whose Ca concentration is very low Ca (8.02 mg/L) and whose Mg concentration is not too high (66.63 mg/L), though it comprised 92.7 % of the cation composition. The highest concentration of 86 mg/L is recorded in a sample of mineralized groundwater, with TDS of 1260 mg/L, that was formed on the contact between limestons and Neogene sediments (PROTIĆ 1995).

In the Pannonian Basin, Mg concentrations in groundwater are found to range from 5.6 mg/L to 65.30 mg/L. Only one sample with a TDS level of 6095 mg/L has magnesium in concentration above MAC, which is the highest concentration recorded in

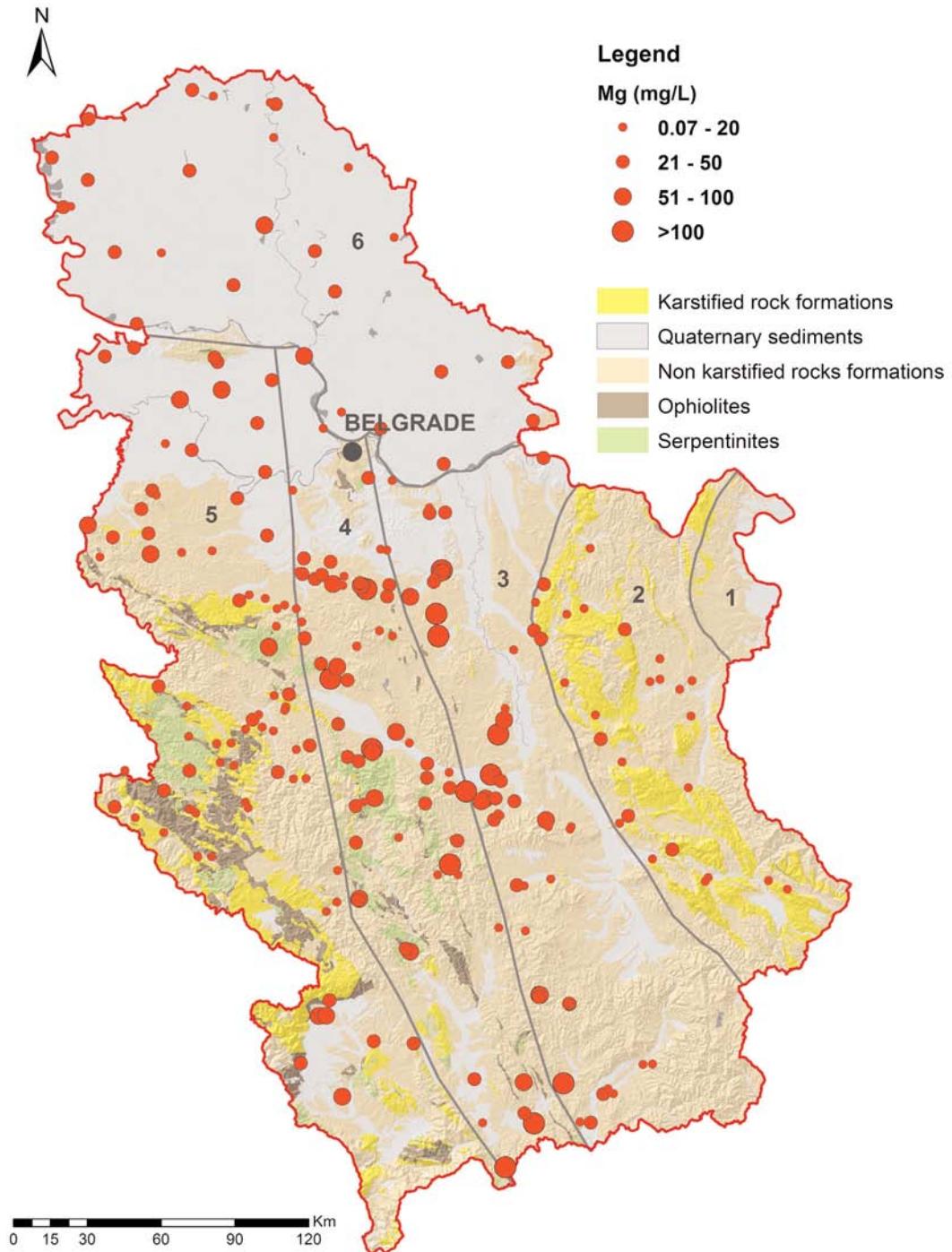


Fig. 4. Distribution of magnesium in Serbia's groundwater resources. Legend: 1, Dacian Basin; 2, Carpatho-Balkanides; 3, Serbian Crystalline Core; 4, Vardar Zone; 5, Inner Dinarides; 6, Pannonian Basin.

the Pannonian Basin (65.30 mg/L). Moderately increased content of Mg on this one groundwater occurrence is a result of the conditions of aquifer forming, at the interface between serpentinites and Tertiary sediments.

Contrary to the previously described provinces, where only a few occurrences of high-TDS groundwater are found to contain high Mg concentrations, the provinces of the Serbian Crystalline Core and

Vardar Zone feature the largest number of occurrences with Mg concentrations exceeding 50 mg/L (MAC for drinking water). In the Serbian Crystalline Core, Mg concentrations are found to range from 0.07 mg/L (the lowest recorded in Serbia) to 183 mg/L, with an average concentration of 40.37 mg/L. Fourteen samples give Mg concentrations above 50 mg/L and they could be divided into two groups: (1) groundwater occurrences whose Mg concentrations are from 50 to 70 mg/L

and whose composition is under the influence of schists, metasandstones and Neogene sediments containing magnesium-rich silicate minerals, and (2) occurrences of high-TDS groundwater (up to 2777 mg/L) associated with schists, whose Mg concentrations measured from 87 to 183 mg/L.

Mg concentrations in the Vardar Zone groundwater are found to be somewhat higher than in the other four provinces. They are ranged from 0.73 mg/L to 324 mg/L. The average is 46.33 mg/L, which is the highest average Mg concentration among the provinces. Two groups of groundwater occurrences in the Vardar Zone can be identified: (1) groundwater occurrences with Mg concentrations up to MAC (50 mg/L), or around to the average Mg concentration for this province, generally tracing to limestones under the influence of flysch formations, schists and Neogene sediments, and (2) 16 groundwater occurrences whose Mg concentrations are in excess of 50 mg/L, where as many as seven samples have Mg concentrations above 100 mg/L, and two samples in excess of 300 mg/L. Such high Mg concentrations are likely a result of the extent of the ophiolitic belt of the Vardar Zone (VASKOVIĆ & MATOVIĆ 2010), as suggested by the rMg/rCa ratio. Vardar Zone ophiolites also influence Mg concentrations of the first group of groundwater occurrences in this province, formed not in ophiolites but other rock formations (primarily karstified Triassic limestones interchanging with less pervious diabase-chert formations and ultramafic rocks).

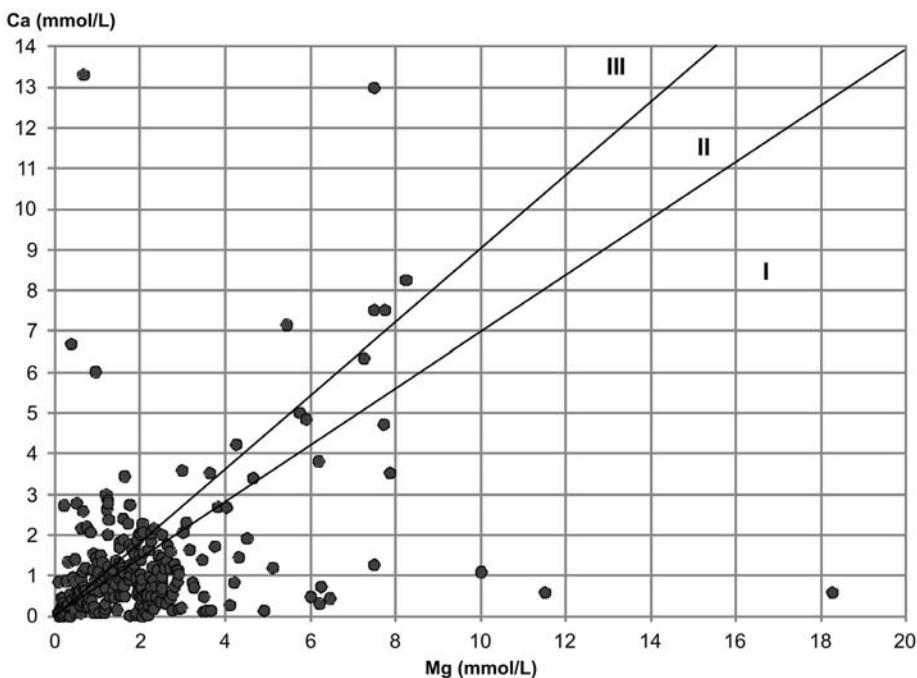


Fig. 5. Magnesium-to-calcium ratio of groundwater. Legend: I, Groundwater formed in limestones; II, Groundwater associated with dolomites and dolomitic limestones; III, Groundwater tracing to magnesium-rich silicate rocks (ophiolites and ultramafic rocks).

### Mg/Ca ratio in groundwater of Serbia

The Mg/Ca ratio (in mequ) of groundwater is very important because it is an indication of the lithological composition of the aquifer matrix. As such, a Mg/Ca ratio of 0.7 suggests that the groundwater was formed in limestones. Groundwater occurrences with an Mg/Ca ratio of 0.7–0.9 are generally associated with dolomitic limestones, while an Mg/Ca ratio greater than 0.9 is indicative of groundwater tracing to Mg-rich silicate rocks. If the ratio is greater than 1, the groundwater traces to ophiolites and ultramafic rocks, as well as ophiolitic detritus in the sediments (MANDEL & SHIFTAN 1981).

The Mg/Ca ratio of the studied groundwater occurrences in Serbia was found to be from 0.01 to 20.30, suggesting diverse lithological compositions and complex geology, or groundwater occurrences tracing to a variety of rocks. Based on the Mg/Ca ratio, 57 % of the groundwater occurrences traced to limestones, only 11 % to dolomites, and 32 % to silicate rocks, of which 25 % to ophiolites and ultramafic rocks (Fig. 5).

The importance and accuracy of classification of the types of rocks in which groundwater is formed, based on the Mg/Ca ratio, is best demonstrated by the Carpatho-Balkanide Province. There, 30 % of the province features karstified rock formations, in which a karst aquifer was formed (STEVANOVIĆ 1991). According to the Mg/Ca ratio, 24 of 26 groundwater samples from this province traced to limestones, as corroborated by the geological makeup.

One sample exhibit Mg/Ca ratio of 0.87, and their chemical composition was under the influence of dolomites in conjunction with limestones. Also, only one sample has ratio above 0.9, suggesting the presence of silicate rocks and corroborated on the ground by andesites and flysch sediments in a portion of this province, along with dominant Cretaceous limestones. This groundwater occurrence is under the influence of silicate rocks but the aquifer is not formed in them, otherwise it would be much higher.

Looking at the calculated Mg/Ca ratios compared with the geological makeup, it becomes apparent that the Mg/Ca ratio of the rock type in which the groundwater occurs matches the geological makeup, such that this ratio can be used to determine the

genesis of groundwater or at least to narrow down the list of possibilities if data on the geological environment of its origin are not available; in other words, this parameter can be used to determine the effect of lithology on the formation of the chemical composition of groundwater.

## Conclusion

It is evident from the results of this research that magnesium concentrations in Serbia's groundwater resources vary over a wide range from 0.07 to 324 mg/L. Uneven distributions and large differences in concentrations have been noted not only between provinces, but also within a single province, as a result of complex geology, attesting to the fact that lithology is the main driver of the chemical composition of groundwater. Additionally, total dissolved solids (TDS) are a very significant parameter as the concentration of this ion, in high-TDS groundwater is considerably higher than in low-TDS groundwater.

Analyses of the Mg/Ca ratio of groundwater in Serbia and the identification of the types of rocks in which groundwater occurs, revealed, based on specified theoretical values, that these ratios largely matched the geological makeup on the ground. It was, therefore, safe to conclude that the Mg/Ca ratio may be used as a parameter for tentative, not definitive, identification of the types of rocks that had a dominant influence on the formation of the chemical composition of groundwater.

The magnesium to calcium ratio is also an important drinking water parameter and the recommended (ideal) ratio of these two ions in water is 1:2. Given that, according to the recommended Mg/Ca ratio more than 60 % of the occurrences of groundwater in Serbia originated in limestones and that quite a few of them exhibit the ideal ratio, such groundwater is precious from a drinking water supply perspective.

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## Резиме

### Дистрибуција магнезијума у подземним водама Србије

Магнезијум је значајан елемент за већину стенских комплекса и битан састојак многих минерала, као што су тамни феромагнезијумски минерали (оливин, пироксени, амфиболи), али и серпентин, талк, бруцит, хлорит, биотит, турмалин, доломит, магнезит, спинел. Садржај Mg у литосфери је различито процењен од 132 до 158 mg/g, а највећи садржаји Mg су у ултрамафитима. У незагађеним, плитким подземним водама садржај магнезијума је од 0,1–1,2 до око 50 mg/L. У процесу распадања стена,  $Mg^{2+}$  улази у хидросферу углавном приликом распадања тамних феромагнезијумских минерала, хлорита, магнезијумских калцита и доломита.

У циљу сагледавања начина дистрибуције магнезијума у подземним водама Србије узети су узорци и анализиране су појаве подземних вода (маломинерализованих и минерализованих) са 253 локација у Србији, формиране у различитим стенским масама (магматске, метаморфне, седиментне стene) и које потичу из различитих типова издани. Како је геологија Србије веома сложена, тешко је посматрати и анализирати целокупну територију одједном, па је ради једноставнијег прегледа, анализирање и тумачење података вршено према извојеним хидрогеолошким рејонима.

На подручју Србије јављају подземне воде различитих типова, температуре као и минерализације, што је последица сложених геолошких услова терена. У анализираним подземним водама минерализација је у опсегу од 29 mg/L до преко 20 g/L. Преовладајући анјон је  $HCO_3^-$ , стога осим неколко појава сулфатних, хлоридних и вода хидрокарбонатно-сулфатног и хидрокарбонатно-хлоридног типа, више од 90 % чине хидрокарбонатне воде. Према катјонском саставу доминирају калцијумске, натријумске или воде мешовитог

типа (Ca-Na, Ca-Mg, Ca-Mg-Na). Четири појаве су магнезијумске воде, са уделом магнезијума већим од 75 %екв.

Концентрације магнезијума у подземним водама Србије се крећу у веома широком опсегу од 0,07 до 378 mg/L, док је средњи садржај 32,10 mg/L. Садржај магнезијума у води зависи од гелошке грађе и тектонике терена у којима су формиране воде, типа воде као и минерализације, јер је код минерализованих вода садржај овог јона већи него код маломинерализованих, иако те воде нису по типу чисто магнезијумске. Од укупног броја анализираних појава, концентрације магнезијума код само 18 %, прелазе максималне дозвољене концентрације према правилнику о квалитету воде за пиће (50 mg/L). Појаве код којих су концентрације Mg преко МДК, су генетски углавном везане за стene попут доломита и доломитичних кречњака, као и за контакт ових стена са флишом, неогеним седиментима, испуцалим и распаднутим пешчари-ма и лапорцима, односно са стенама које у свом саставу садрже магнезијумске минерале. Повише-ни садржај Mg је детектован и у водама са веома повишеном минерализацијом, најчешће везаним за шкриљце. Концентрације Mg преко 70 mg/L су забележене код вода везаних за серпентините, испуцале харцбургите, контакте доломита и сер-пентинита.

Дистрибуција магнезијума у подземним водама Србије није равномерна, али се одређене законо-мерности могу уочити. Неравномерна дистрибуција и велике разлике у концентрацијама могу уочити, не само између различитих рејона, већ и у оквиру једног посматраног рејона, што је последица сложене геолошке грађе терена и доказ да је утицај литологије на формирање хемијског састава подземних вода главни фактор. Поред тога, минерализација воде је веома значајан параметар, јер су концентрације магнезијума у водама високе минерализације значајно веће него у маломине-рализованим водама.

Однос  $rMg/rCa$  у подземним водама је веома важан јер указује на литолошки састав стена у којима су формиране подземне воде. Однос  $rMg/rCa$  у анализираним подземним водама у Србији је у опсегу од 0,01 до 20,30 што указује на разноврсан литолошки састав и сложену геолошку грађу, односно на присуство вода које потичу из различитих стенских маса. На основу односа  $rMg/rCa$ , 58 % појава подземних воде потиче из кречњака, само 9 % из доломита, док 33 % појава су воде везане за силикатне стene, од којих 25 % за олиолите и ултрамафите.