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Editor:
Lidia Razowska-Jaworek

Calcium and Magnesium in Groundwater

Occurrence and Significance for Human Health

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Chapter 3

Hydrogeochemical distribution of Ca and Mg in groundwater in Serbia

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ABSTRACT

Ca and Mg are chemical elements commonly found in the environment and the main constituents of many types of minerals and rocks. They are also essential to Man. Owing to their abundance in nature, they are present in all water resources and generally occur as the dominant cations with 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 Ca and Mg. Ca and Mg concentrations in groundwater in Serbia vary over a wide range and their distribution is not uniform. The variation in the concentrations of these ions depends on the hydrogeological province, while in any 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 structure is much more complex than that of the Carpatho-Balkanides.

3.1 INTRODUCTION

Ca and Mg are lithophile elements that play an important role in the composition of groundwater and in the environment in general, and are also essential to the human body (Jovic & Jovanovic, 2004). Ca is the most abundant alkaline earth metal in the Earth's crust and the main ingredient of many minerals and rocks. The most common Ca-rich minerals are: calcite, aragonite, dolomite, gypsum, apatite, clinopyroxenes, plagioclases, hornblende and epidote (Clare & Rhodes, 1999). According to estimates, the lithosphere contains 16.2–19.3 mg-Ca/g. The highest concentrations have been recorded in carbonate rocks (limestones, dolomites), and basalts among igneous rocks (Hitchon *et al.,* 1999). As rocks weather, Ca dissolves readily and enters the hydrosphere. The carbonate equilibrium is the main driver that restricts Ca migration in natural waters. In solution, Ca occurs as a bivalent ion, Ca^{2+} (Hitchon *et al.,* 1999).

Mg is a significant component of most rock formations and an important ingredient in many petrogenic minerals, such as dark ferromagnesian minerals (olivine, pyroxenes, amphiboles), but also minerals like serpentinite, talc, brucite, chlorite, biotite, tourmaline, dolomite, magnesite and spinel (Jovic & Jovanovic, 2004). 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 a sedimentary environment, Mg largely occurs in association with the carbonate ion, predominantly as dolomite $CaMg(CO_3)_2$. As a result of rock weathering, Mg^{2+} usually enters the hydrosphere as dark ferromagnesian minerals (e.g. chlorite, Mg-calcite and dolomite) degrade. In unpolluted shallow groundwater, Mg concentrations range from 0.1–1.2 to about 50 mg/l (Cox, 1995).

Ca and Mg are essential elements to Man and their impact on human health is enormous. Ca is found in the human body more than any other mineral. It takes part in the formation of bones and teeth. Muscle activity and the transmission of nerve impulses rely on Ca. It is valuable in blood coagulation, cardiac activity and enzyme production. In conjunction with Mg, it supports the function of the human heart. Lack of Ca increases the risk of high blood pressure and heart failure, while a prolonged deficit may lead to osteoporosis. Mg plays a multiple role in the human body: it acts directly on the neuromuscular plate, is essential for normal vitamin C and vitamin B_1 activity, takes part in enzymatic processes leading to energy production, reduces coagulation levels, protects the inner walls of blood vessels from fibrosis, and catalyses the utilisation of fats, proteins and carbohydrates (Teofilovic *et al.*, 1999; Cotruvo *et al.*, 2009).

Serbia is a country located at the crossroads of Central and South East Europe, covering the southern part of the Pannonian Plain and the central Balkans, lying between a massif and the Carpathian Mountains in the east, the Dinaric Alps in the west, and the Morava Valley – an intersection of land routes which lead southwards, towards Salonika, and eastwards, towards Asia Minor. The country is landlocked and borders Hungary to the north; Romania and Bulgaria to the east; Macedonia to the south; and Croatia, Bosnia, and Montenegro to the west; it also borders Albania through the disputed region of Kosovo. The capital of Serbia, Belgrade, is among Europe's oldest cities and one of the largest in East Central Europe.

The geology of Serbia is highly complex and not conducive to generalised assessments. Broad geological provinces have been identified based on geotectonic units. In general terms, the provinces are (Filipovic *et al.*, 2005): the Carpatho-Balkanides, the Serbian Crystalline Core, the Vardar Zone, the Inner Dinarides and the Pannonian Basin (Figure 3.1).

3.2 APPROACH AND METHOD

The data used in this research were derived from investigations conducted between 2008 and 2012. Groundwater was sampled at 257 locations across Serbia, including groundwater resources featuring low and high Total Dissolved Solids (TDS). The sampling network was designed to cover the entire territory of Serbia evenly 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, no. 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. Ca and Mg concentrations were determined by the ICP-OES method.

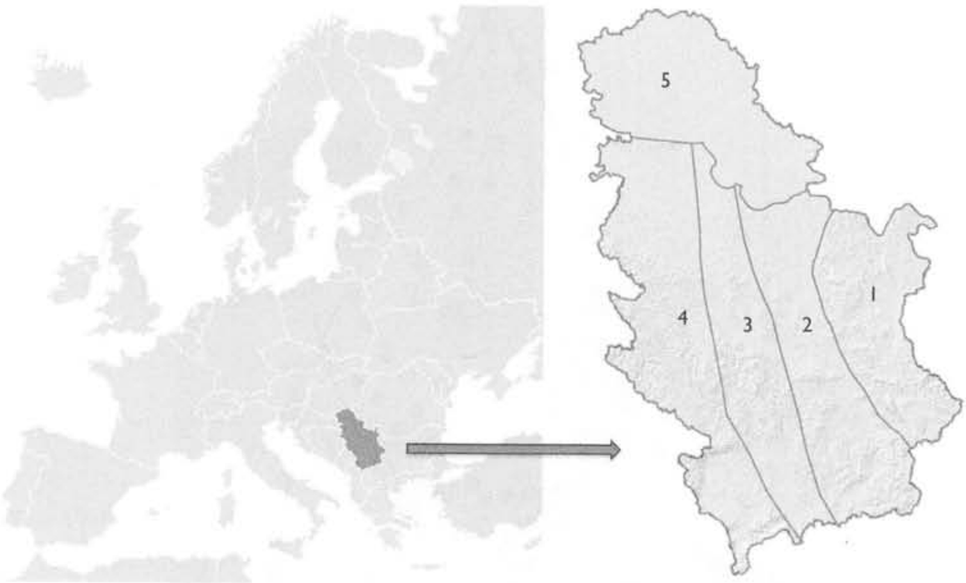


Figure 3.1 Geographic location and hydrogeological provinces of Serbia. Legend: 1. Carpatho-Balkanides; 2. Serbian Crystalline Core; 3. Vardar Zone; 4. Inner Dinarides; 5. Pannonian Basin.

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

3.3 RESULTS AND DISCUSSION

3.3.1 General groundwater quality

Serbia's highly complex geology has resulted in groundwater resources featuring different types, temperatures and TDS levels. The dominant anion was the hydrocarbonate ion. Apart from several occurrences of sulfate, chloride, hydrocarbonate-sulfate and hydrocarbonate-chloride types, more than 90% were found to be of the hydrocarbonate type of groundwater. Of all the samples, three belonged to the sulfate type and two to the chloride group with a chloride portion of 97% equivalent. The latter two occurrences show high TDS levels, in excess of 6000 mg/l. Based on their cation composition, the samples predominantly reflected Ca, Na and composite (Ca–Na, Ca–Mg, Ca–Mg–Na) types of groundwater. Four samples were of the Mg type, with a Mg component in excess of 75% equivalent (Figure 3.2).

With regard to Total Dissolved Solids (TDS), the samples exhibited considerable diversity: from low levels (only 29 mg/l) to very high levels (in excess of 20 000 mg/l).

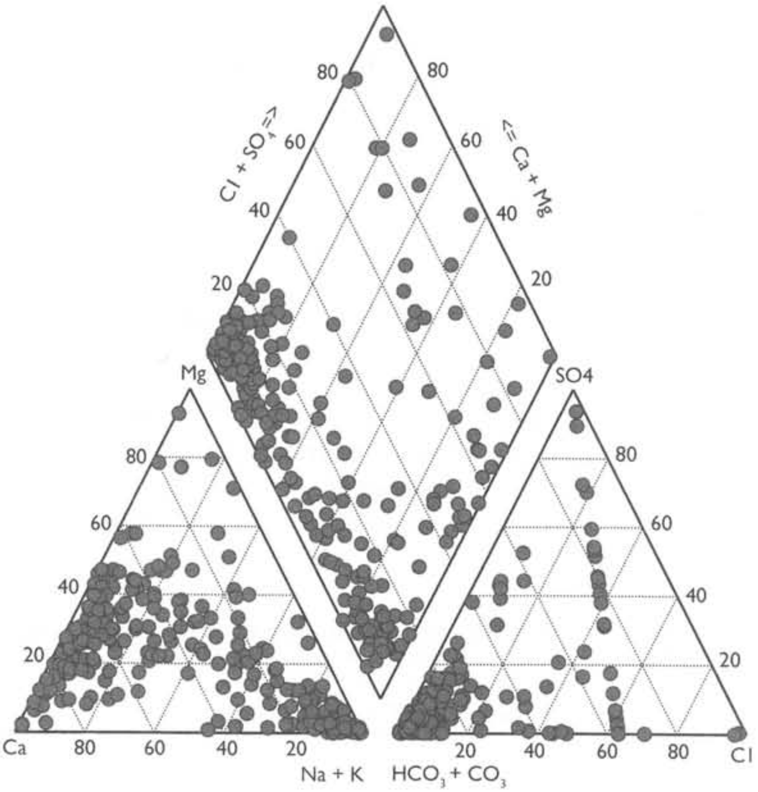


Figure 3.2 Piper diagram of the chemical composition of groundwater samples.

3.3.2 Calcium in the groundwaters of Serbia

Ca concentrations in Serbia's groundwaters were found to vary over a broad range, depending on the type of groundwater and TDS: from 0.60 mg/l to 392.80 mg/l. The median was 82.32 mg/l.

Ninety-five or 37% of the samples had Ca concentrations up to 50 mg/l, while the majority of the samples, 107 or 42%, contained Ca in concentrations ranging from 51 to 100 mg/l, meaning that some 80% of samples featured Ca in concentrations up to 100 mg/l (Figure 3.3).

Of the 257 occurrences groundwater samples, only 20 (8%) featured Ca concentrations in excess of the Maximum Allowable Concentration (MAC), which is 200 mg/l according to national drinking water standards (Figure 3.3). In view of the TDS levels, high Ca concentrations are common, especially in carbonated groundwater, given that elevated CO_2 concentrations enhance the solubility of Ca carbonates and, consequently, increase Ca concentrations in the groundwater.

As Ca is abundant in the Earth's crust, it is found in all natural water resources. Even though Ca^{2+} mostly relates to sedimentary rocks (limestone, dolomite), it is difficult to explain that high Ca^{2+} concentrations have been recorded in groundwater

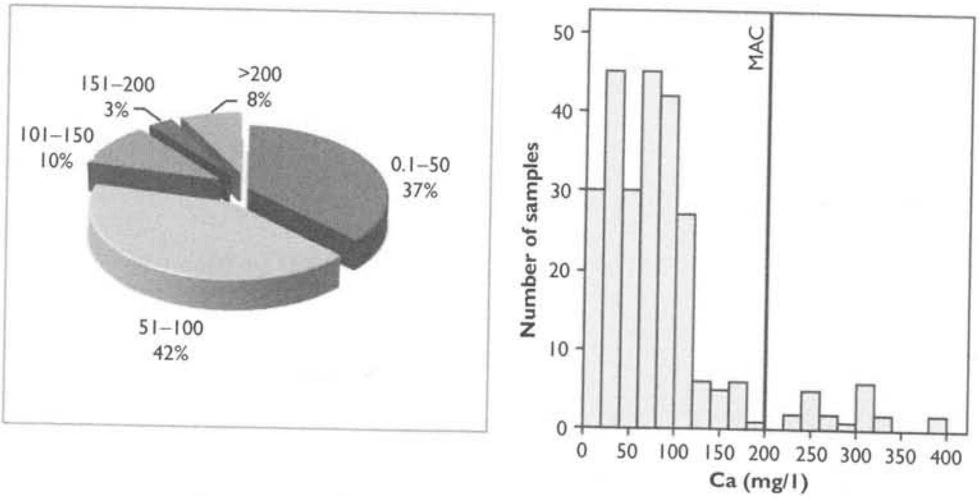


Figure 3.3 Plot of relative frequency of Ca and histogram of Ca concentrations in Serbia's groundwaters.

occurring in limestones because Ca concentrations in groundwater are limited by the solubility of calcium-carbonates. High Ca concentrations in water, typical of mineral water, are conditional upon the presence of other components (CO_2 and other ions), which affect the concentrations of dissolved mineral substances that together contribute to the TDS level of the water.

Ca is not evenly distributed in Serbia's groundwater resources but certain relationships apply with the respective geological makeup and are apparent in the various hydrogeological provinces (Figure 3.4).

For example, the most uniform distribution of Ca in groundwater is found in the Carpatho-Balkanides, which is as expected given that groundwater in this province occurs in predominant karstified rock formations. Ca concentrations are typical of the low-TDS HCO_3–Ca type of groundwater, with an average of about 75 mg/l. One sample had a Ca concentration of 240 mg/l; this was carbonated groundwater with a TDS level of 1294 mg/l, whose origin is rather complex and governed by tectonics and the geological makeup of the rocks underlying the karstified formations. The presence of a hidden granite intrusion has caused the generation of CO_2, enhanced the solubility of calcium-carbonates and enriched this groundwater with Ca.

The Inner Dinarides of western Serbia also feature certain regularities in the distribution of calcium. Groundwater occurrences originally associated with karstified formations were largely found to be of the low-TDS HCO_3–Ca type, with calcium concentrations up to 70 mg/l. Contrary to this type, high-TDS groundwater occurrences whose calcium concentrations measured up to 380 mg/l were associated with flysch sediments and schists. The origin of such groundwater is intricate and tectonics plays an important role in the formation of its chemical composition.

Within the Pannonian Basin, groundwater is of the hydrocarbonate type, with hydrocarbonate ion concentrations in excess of 50% equivalent. Their cation

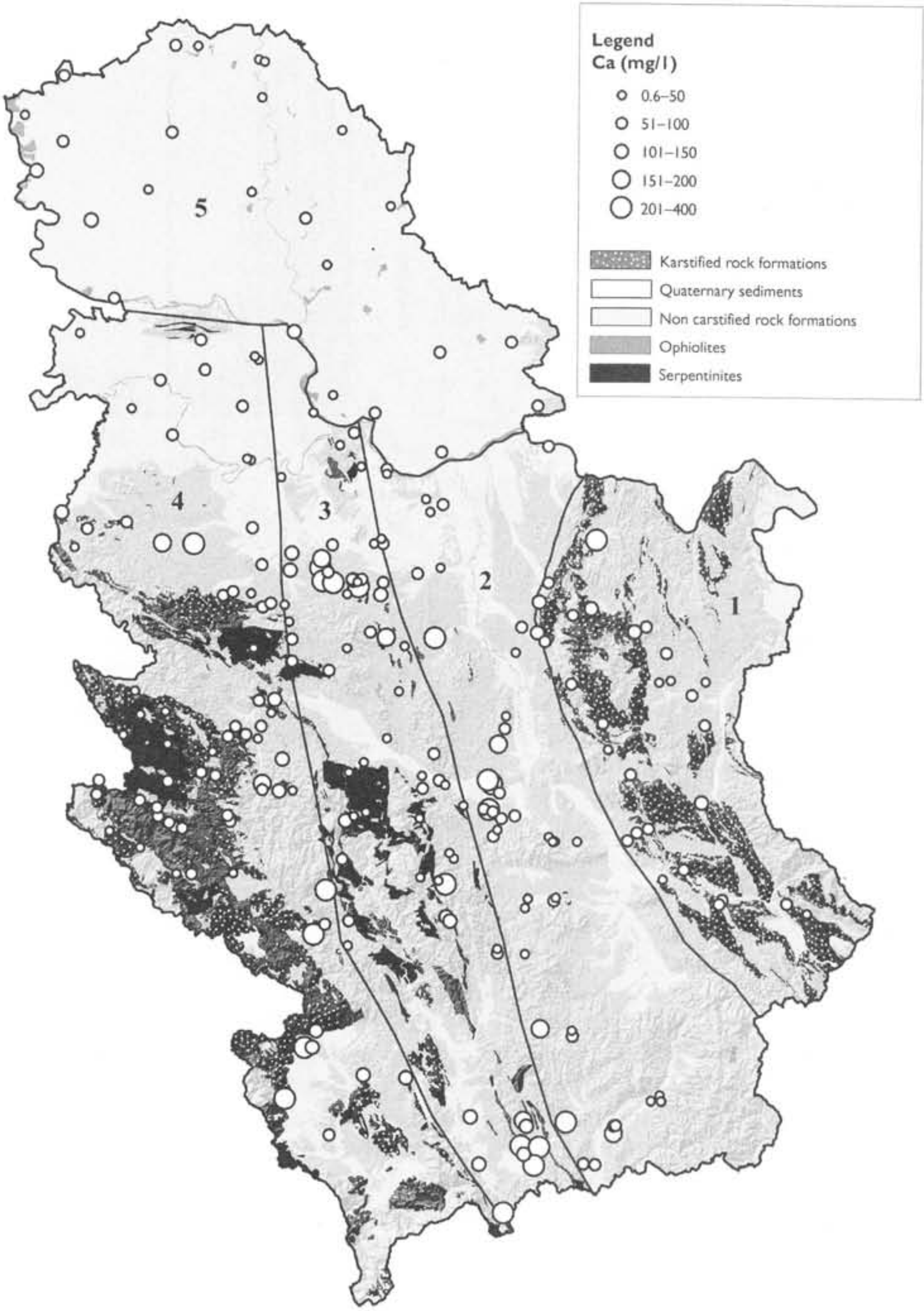


Figure 3.4 Ca distribution in Serbia's groundwaters. Legend: 1. Carpatho-Balkanides; 2. Serbian Crystalline Core; 3. Vardar Zone; 4. Inner Dinarides; 5. Pannonian Basin.

Table 3.1 Ca concentrations in different hydrogeological provinces (mg/l).

Province	*Number of samples*	*Minimum*	*Average*	*Maximum*
Carpatho-Balkanides	27	14.00	75.86	240.00
Serbian Crystalline Core	54	0.60	61.22	310.00
Vardar Zone	62	3.00	67.02	392.80
Inner Dinarides	85	1.80	66.66	380.00
Pannonian Basin	29	2.40	53.25	138.00

composition is complex, dominated by Ca, Mg and Na ions in different ratios. Table 3.1 shows that the concentration range in this province is the smallest. Higher concentrations were noted in certain high-TDS samples, such as the occurrence of the Cl–Na type of groundwater with a TDS level of 6600 mg/l, where the Ca concentration was 114.9 mg/l but it constituted only 0.7% of the cation composition.

The largest variations in Ca concentrations were noted in the Serbian Crystalline Core and the Vardar Zone, as a result of the complex geology of Central Serbia. In these provinces concentrations ranged from only 0.6 mg/l to as much as 392.8 mg/l (the highest recorded concentration). This groundwater is of the HCO_3–Mg–Ca–Na type, with a TDS level of nearly 5 g/l.

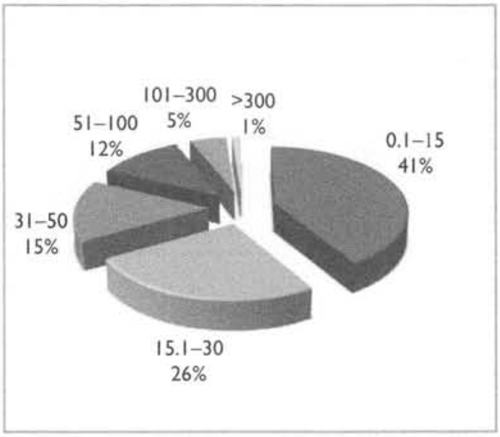
3.3.3 Magnesium in the groundwaters of Serbia

Mg concentrations in Serbia's groundwaters vary considerably, from 0.05 mg/l to 378 mg/l. The average is 34.27 mg/l. Mg concentrations depend on the geology and tectonics, 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, and that this groundwater is not of a pure Mg type.

In most of the samples (106 or 41%), Mg concentrations were only up to 15 mg/l; 26% or 66 samples contained 15 to 30 mg/l, while 15% (39 samples) were between 30 to 50 mg/l. This means that 82% of the groundwater samples collected across Serbia had Mg concentrations up to 50 mg/l, which is the maximum allowable concentration according to national drinking water standards.

Of the 257 samples, 45 samples (or 18%) had Mg concentrations in excess of drinking water standards (Figure 3.5). Compared with Ca concentrations (only 20 of 257 samples exceeding the MAC), Mg had a larger number of exceedances but in view of the total number of samples analysed and the fact that the MAC for Mg is considerably lower than that for Ca, such a proportion was expected.

Mg concentrations in excess of 50 mg/l were found in high-TDS groundwater of up to 6000 mg/l. Such groundwater is generally traced to schists. In several samples of low-TDS groundwater, Mg concentrations measured 50–60 mg/l; such groundwater is largely originally associated with dolomite and dolomitic limestone, as well as interfaces of these rocks with flysch, Neogene sediments, fractured and degraded sandstones and marls, or rocks whose composition includes Mg-rich minerals.



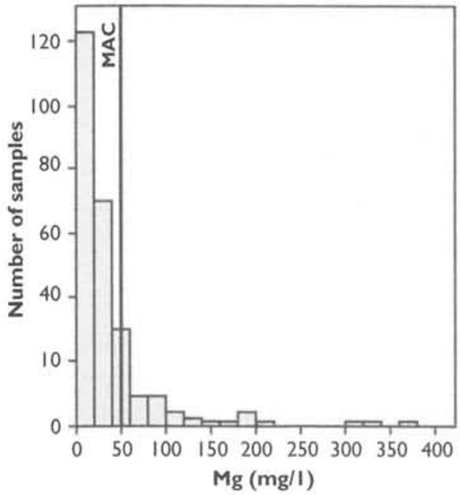


Figure 3.5 Plot of relative frequency of Mg and histogram of Mg concentrations in Serbia's groundwaters.

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 interfaces.

As with Ca, the distribution of Mg is not uniform (Figure 3.6), but regularities resulting from the geological makeup are much more apparent.

Table 3.2 shows the smallest variations but also the lowest Mg concentrations in the Carpatho-Balkanide Province, where the average Mg concentration was found to be 16.52 mg/l. One sample from this province exceeded the MAC (52 mg/l), but this was a result of the influence of flysch in the vicinity, even though the province is dominated by limestone and dolomitic karstified rock formations, as corroborated by the Mg to Ca ratio.

The Inner Dinarides of western Serbia contained an average Mg concentration in groundwater of 25.29 mg/l; the concentrations of this ion were generally below the MAC, although there were several exceptions where concentrations were considerably higher (up to 378 mg/l). The occurrences that gave Mg concentrations up to the average level (about 25 mg/l) were generally low-TDS groundwater resources from 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 were recorded in samples collected from groundwater occurrences in limestones influenced by serpentinites, harzburgites and similar rocks that make up the Dinaride ophiolite zone. This province featured a distinct occurrence of groundwater of the Mg type associated with pure serpentinites and harzburgites. This was low-TDS groundwater (350 mg/l), in which the Ca concentration was very low Ca (8.02 mg/l) and the Mg concentration was not high (66.63 mg/l), though it comprised 92.7% of the cation composition. The highest concentration of 378 mg/l was recorded in a sample that

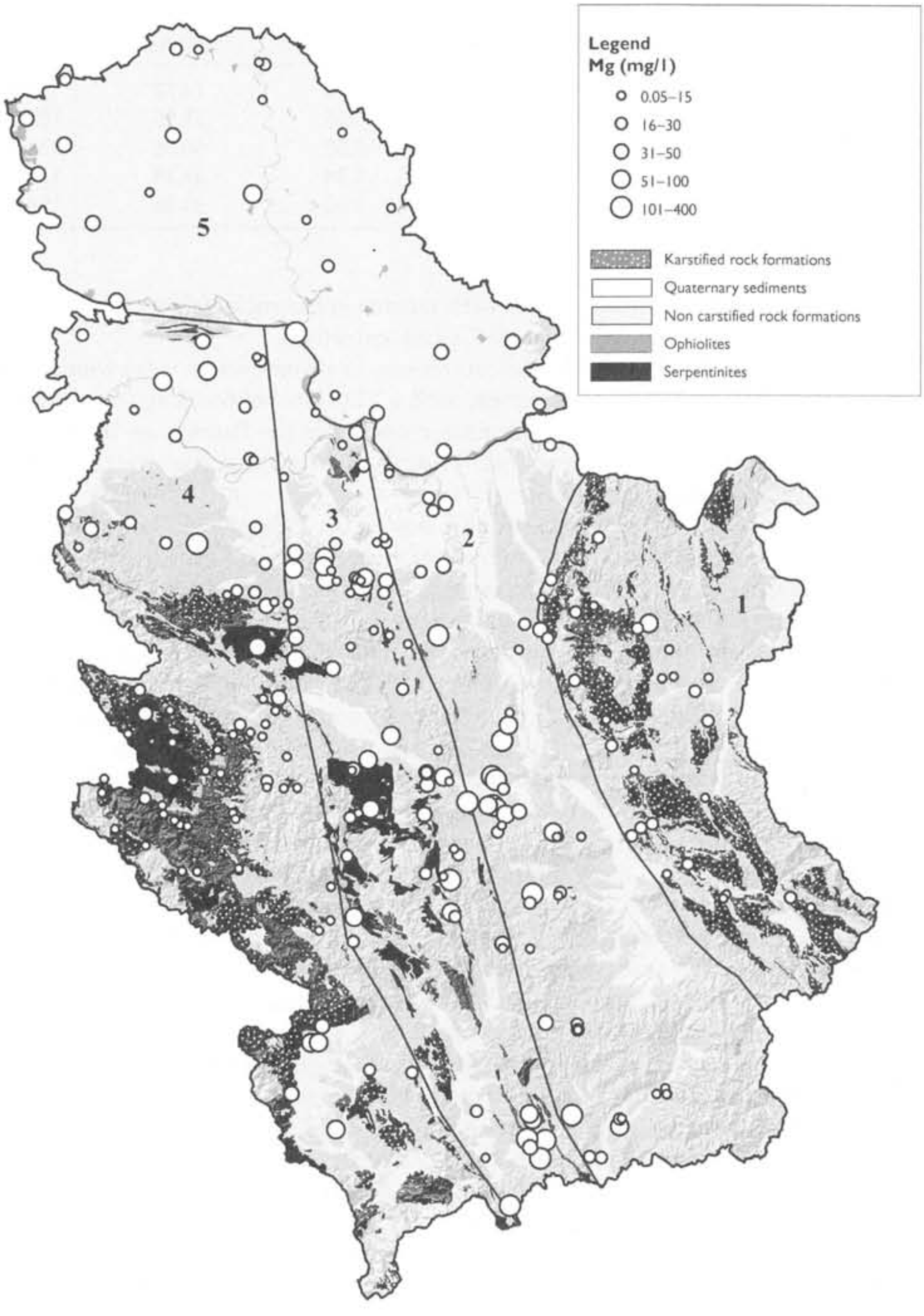


Figure 3.6 Distribution of magnesium in Serbia's groundwaters. Legend: 1. Carpatho-Balkanides; 2. Serbian Crystalline Core; 3. Vardar Zone; 4. Inner Dinarides; 5. Pannonian Basin.

Table 3.2 Mg concentrations in different hydrogeological provinces (mg/l).

Province	*Number of samples*	*Minimum*	*Average*	*Maximum*
Carpatho-Balkanides	27	1.00	16.52	52.00
Serbian Crystalline Core	54	0.05	39.95	183.00
Vardar Zone	62	0.60	50.06	324.00
Inner Dinarides	85	3.24	25.29	378.00
Pannonian Basin	29	5.60	33.36	194.00

had a TDS of 4995 mg/l, associated with metamorphic rocks (slates, phyllites) and sandstones, which also had the highest Ca concentration.

In the Pannonian Basin, Mg concentrations in groundwater were found in the range 5.6 mg/l to 50.5 mg/l. One sample with a TDS level of 6620 mg/l had 194 mg-Mg/l, which was the highest concentration recorded in the Pannonian Basin. Such a high Mg concentration was a result of the aquifer formation at the interface between serpentinites and Tertiary sediments.

Unlike the other provinces, where only a few occurrences of high-TDS groundwater were found to contain high Mg concentrations, the provinces of the Serbian Crystalline Core and Vardar Zone have the largest number of occurrences with Mg concentrations exceeding 50 mg/l (MAC for drinking water). In the Serbian Crystalline Core, Mg concentrations were found to range from 0.05 mg/l (the lowest recorded in Serbia) to 183 mg/l, with an average concentration of 39.95 mg/l. Fourteen samples had Mg concentrations above 50 mg/l and they could be divided into two groups: (1) groundwater occurrences with Mg concentrations are between 53 and 70 mg/l and whose composition is influenced by schists, metasandstones and Neogene sediments containing Mg-rich silicate minerals, and (2) occurrences of high-TDS groundwater (1523 to 4995 mg/l) associated with schists, whose Mg concentrations measured from 80 to 183 mg/l.

Mg concentrations in the Vardar Zone groundwater were found to be higher than in the other four provinces. They ranged from 0.6 mg/l to 324 mg/l. The average was 50.06 mg/l, which was 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 up to the average Mg concentration for this province, generally related to limestones and influence by flysch formations, schists and Neogene sediments, and (2) 20 groundwater occurrences whose Mg concentrations were in excess of 50 mg/l, where as many as seven samples had Mg concentrations above 100 mg/l, and two samples were 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, 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).

3.3.4 Mg/Ca ratio in groundwater of Serbia

The Mg/Ca ratio of groundwater is important because it is an indicator of the lithological composition of the aquifer matrix. For instance, a Mg/Ca ratio of 0.7 may

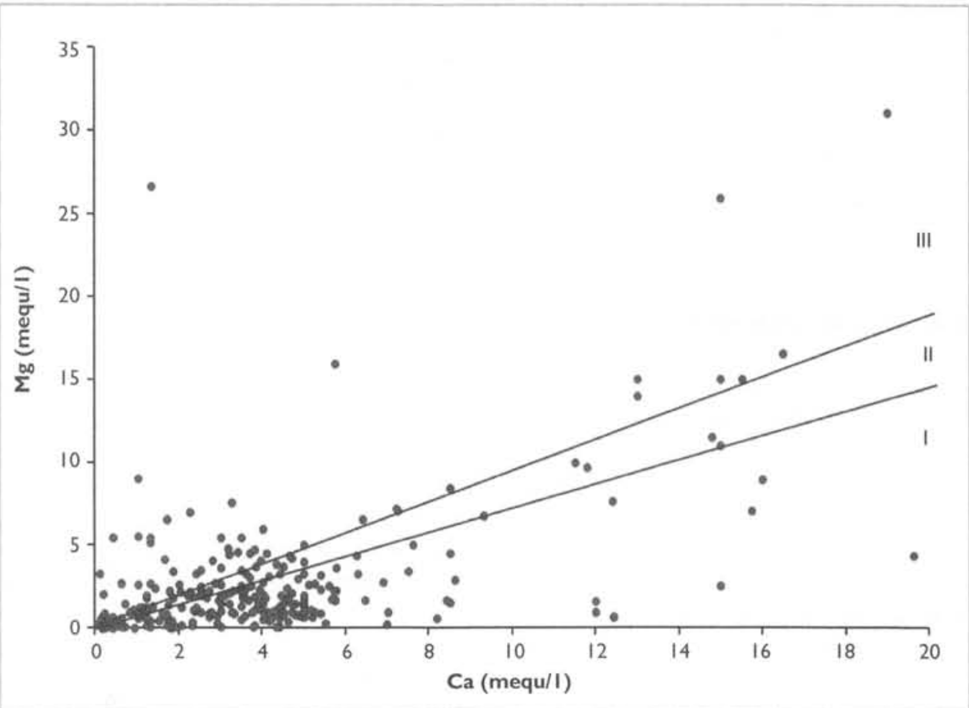


Figure 3.7 Mg/Ca ratio of groundwater. Legend: I. Groundwater formed in limestones; II. Groundwater associated with dolomites and dolomitic limestones; III. Groundwater tracing to Mg-rich silicate rocks (ophiolites and ultramafic rocks).

suggest that 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 from Mg-rich silicate rocks. If the ratio is greater than 1, the groundwater relates to ophiolites and ultramafic rocks, as well as ophiolitic detritus in the sediments (Mandel & Shiftan, 1981).

The Mg/Ca ratio of the groundwater samples in Serbia was found to range from 0.02 to 36.62, suggesting diverse lithological compositions and complex geology, or groundwater occurrences relating to a variety of rock types. Based on the Mg/Ca ratio, 58% of the groundwater occurrences related to limestones, only 9% to dolomites, and 33% to silicate rocks, i.e. 25% to ophiolites and ultramafic rocks (Figure 3.7).

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 feature karstified rock formations, and karst aquifers. According to the Mg/Ca ratio, 24 out of the 27 groundwater samples from this province related to limestones, as corroborated by the geological makeup. Two samples had Mg/Ca ratios of 0.7 and 0.9, and their chemical composition was under the influence of dolomites in conjunction with limestones. Only one sample had a ratio of 0.91, suggesting the presence of silicate rocks and corroborated on the ground by andesites and flysch sediments in part of this province, along with dominant Cretaceous limestones. This groundwater is influenced by silicate rocks but the aquifer was not contained in them, such that the ratio is close to the limit of 0.9 for this group. Otherwise it would be much higher.

Looking at the calculated Mg/Ca ratios compared with the geological makeup, it is apparent that the Mg/Ca ratio of the rock type in which the groundwater occurs matches the geological makeup, and that this ratio can be used to determine the origin of groundwater or at least to narrow down the list of possibilities if data on the geological environment are not available; this parameter can be used to determine the effect of lithology on the formation of the chemical composition of groundwater.

3.4 CONCLUSION

It is evident that Ca and Mg concentrations in Serbia's groundwaters vary over a wide range: 0.6 to 392.8 mg/l for Ca and 0.05 to 378 mg/l for Mg. 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 the groundwater. Additionally, Total Dissolved Solids (TDS) are a significant parameter as the concentration of one or both of these ions 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. 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 the groundwater.

The Mg/Ca 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 groundwaters in Serbia originate from limestones and that quite a few of them exhibit the ideal ratio, but such groundwater is precious from a drinking water perspective.

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INTERNATIONAL ASSOCIATION OF HYDROGEOLOGISTS SELECTED PAPERS

Calcium and magnesium are abundant in groundwater, but the role of groundwater as the essential source of these important nutrients is very often neglected. Hydrogeochemical studies have focused mainly on the distribution and behaviour of constituents that cause deterioration of water quality, such as: nitrate, nitrite or iron and manganese. Therefore, most recent books and papers concentrate mainly on these constituents and only a small number of papers describe the results of groundwater studies on other important water components such as calcium or magnesium. Calcium and magnesium are of great importance to human wellbeing and inadequate intake of either nutrient can impair health. The main objective of this book is to present a selection of studies on calcium and magnesium in groundwater and to highlight their importance for human health. The chapters in this book are written by hydrogeologists and hydrochemists as well as medical doctors and include: methods of investigation of calcium and magnesium in groundwaters, distribution and behaviour of calcium and magnesium in different aquifers, calcium and magnesium in bottled mineral and spring waters, and the significance of calcium and magnesium in waters for human health.

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