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МАКЕДОНСКО ГЕОЛОШКО ДРУШТВО

ВТОР КОНГРЕС
на
Геолозите на Република Македонија

ЗБОРНИК НА ТРУДОВИ

Уредници:
Јовановски, М. & Боев, Б

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HYDROGEN ISOTOPIC STUDY OF THE BOROVIĆ MINERALIZED SYSTEM, KRATOVO-ZLETOVO VOLCANIC AREA

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Abstract
Wide zones of intensive hydrothermal alterations were determined at the Borović area. Silicification, limonitization, kaolinization etc., are the progressive alterations with distinctive occurrences at the surface. Stockwork veins and veinlets are dominant too. Detailed studies have shown that this system at the Borović has been mineralized, where at few randomly taken samples from the silicificied zones has been determined presence of gold within the range of 0.10 up to 0.62 g/t Au, followed by increased concentrations of copper, silver, molybdenum etc. Also, in this mineralized system have been determined presence of geothermal mineralized waters with temperatures up to 50ºC and artesian flow of about Q=10 l/s within the Povisica river flow. Isotopic studies of hydrogen and oxygen or ratios H/D and $\delta^{18}O$ in samples taken from thermomineral waters from wells in the Borović locality have shown values within the range -10.62÷ -8.61 $\delta^{18}O$ and -72.70÷ -56.90 δD, which points out that the origin of these mineralized waters is mainly of meteoric character.. Namely, there were slight discrepancies from the MWL (Meteoric Water Line), which reflects the direct fractionation of O and H isotopes in meteoric water at the contact with adjacent rocks and deep hydrothermal fluxes

Key words: Isotopes, alterations, mineralization, hydrogeochemistry, Borović

INTRODUCTION

Locality Borović in the geological literature is known since the first half of the twentieth century when the concession for geological exploration of this area had English company Selected Mines Ltd. However, more specific data from the studies of the Kratovo-Zletovo volcanic area and of course the Borović itself can be found in works of Pantic et al., (1972), Klajn (1977), Serafimovski (1990), Bogoevski (1998) etc. Later on this locality has been explored as potential locality for geothermal energy. In that direction we wouls like to stress out the works of Rakic et al. (1993), Rakic et al. (1996), Gorgieva (1995), Serafimovski et al. (2001), Rakic and Stolic (2002), Ristova (2011), Verbovšek et al., (2011) etc.

All the explorations and data contained within numerous materials are pointing out that in the vicinity of Borović has been determined copper and gold mineralizations, as well as significant quantities of thermomineral waters, which exploitation is of continuous character. This was our reason in this paper to review the Borović mineralized system, its alterations and H/D and O isotopes within its thermomineral waters.

GEOLOGY OF THE STUDIED AREA

Studied area lies below the Borović hill (651 m), few kilometers west from town of Kratovo in NE Macedonia (42.07ºN, 22.08 ºE). The nearest villages are Topolović and Turalevo in the north and Filipovci in the south. Area belongs to Zdravevci geothermal system, named after a nearby smaller village. The region is also known as Borović (or Borovik, originally Боровиќ) ore field, and is therefore noted further on in the paper as Zdravevci-Borović geothermal system. Borović is one of the volcanic manifestations in the wider Kratovo-Zletovo volcanic area, and several wells were drilled in the valley of river Povišnica (Povišica) in late 1980's.

Kratovo-Zletovo area lies in a former volcanic crater or caldera (Turalevo crater), which was later significantly altered by intensive hydrothermal alternation, silicification (up to 99% SiO_2), jarositization and kaolinitization. The area was intensively studied for the occurrence of ore minerals, and high values of Pb, Zn and Cu were found. Area was mostly studied by (Klajn, 1977) and (Rakić et al., 1993).

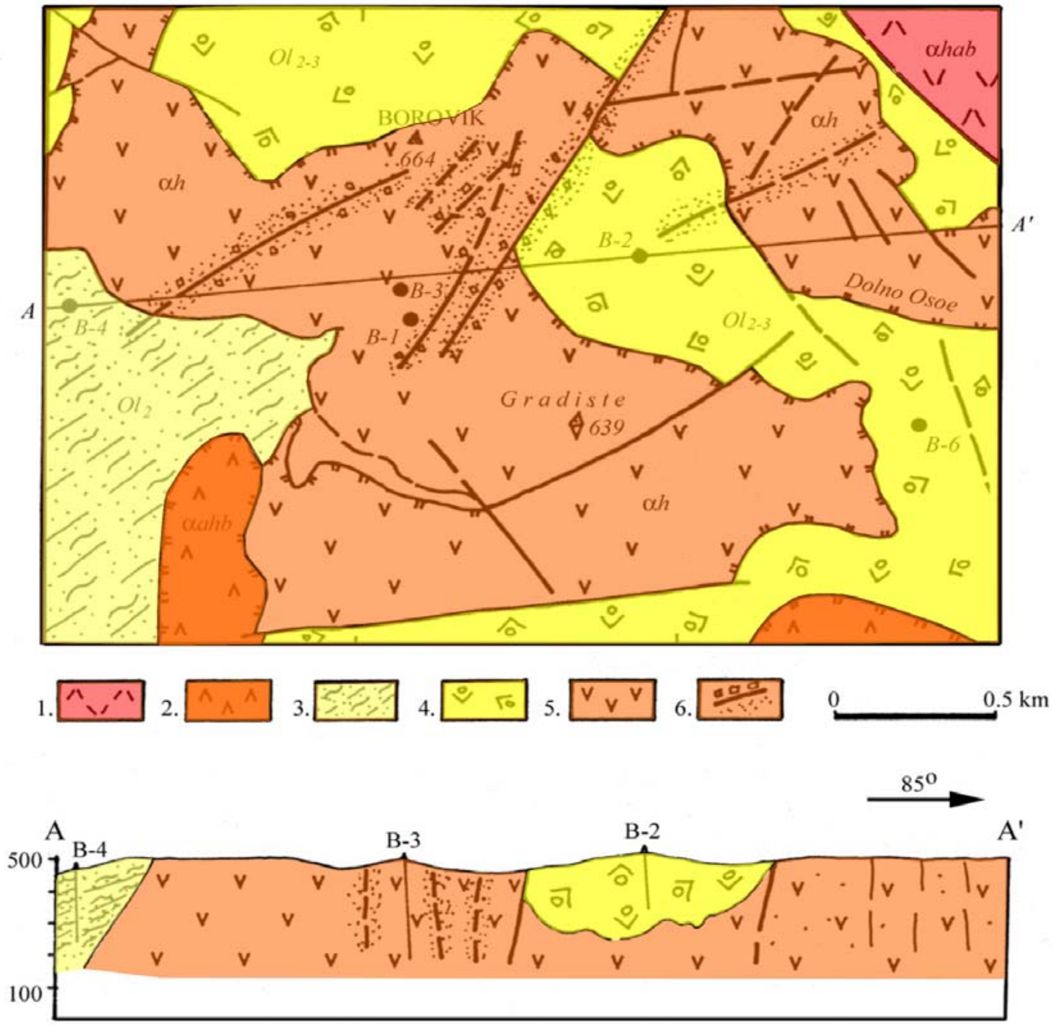

Figure 1. Geological map and cross section through the Borovik mineralized system (Serafimovski, 1990) 1. Hornblende-augite-biotitic andesite; 2. Augite-hornblende-biotitic andesite; 3. Tuffaceous sandstones; 4. Tuffaceous sandstones, breccia and marls; 5. Hornblende andesite; 6. Faults (often mineralized)

Mineralized area Bukovik is located in the NW parts of the Kratovo-Zletovo ore district, between the Filipovci and Turalevo villages. The terrain is of low mountanious nature with average altitudes of 450 to 750 m and few representative hills such are: Gradiste, Borovik, Golak and some other with extensive erosion.

Geological setting is heterogeneous and in generals consists of volcanic materials, tuffaceous breccias, sandstones, marls etc. Oligocene tuffaceous breccias, sandstones and marls have been intruded by biotite-augite andesites, hornblende andesites and sometimes occur even younger volcanic outburst as smaller or larger plate-like forms.

Structural setting is quite similar to the other parts of the adjacent vicinity of this mineralized area, within the Kratovo-Zletovo ore district. Namely, the major fault and structural systems are of NE-SW and NW-SE direction and rarely of E-W direction, which at the crater Turalevo and Borovik intersect each other and in such a manner represent an area suitable for deposition of ore mineralization. The fault structures are related with intensive hydrothermal alterations and post-volcanic manifestations. In regards of hydrothermal alterations the most abundant one is the silicification, followed by kaolinitization, sericitization, chloritization, pyritization etc.

Up to date data have shown that within the Borovik mineralized system are present numerous hydrothermally altered zones that occasionally are mineralized, mainly with copper and gold mineralization and sporadically with lead and zinc. All the facts are pointing out that the major focus should be given to the exploration of copper mineralizations.

All the occurrences of copper mineralization according to the host rocks where they have located can be divided into: pyrite occurrences with chalcopyrite in tuffaceous series, which have been intensively altered (with notable presence of argillites and jarosites). There have been confirmed "iron hats" (Klajn, 1977). Especially characteristic is pyritization with "iron hats" along left tributary of the Povisica river southern of Borovik. Sampling of adits and exploration drill holes have shown presence of copper up to 0.1% Cu and plenty of pyrite, reaching up to 20% of mineralized mass. The most copper is related to the crystal lattices of pyrite either it sporadically occurs in form of chalcopyrite disseminated in pyrite or as very fine individual grains.

Newer detailed explorations (drilled few exploration drill holes) have shown copper and gold mineralization, but however the data are still of limited character and intensity of exploration is low. Preliminary findings are that the mineralization is of quite similar character as it is the one at the Plavica locality and is more intensive around the zones of tectonic crushing. Also, the aforementioned findings indicated that the gold mineralization could be attributed to the existence of epithermal gold deposit of so-called acid-sulfate type.

Gold concentrations were always higher within intensively silicified zones, which are situated along faulting structures or zones of their intersection. At the following table (Table 1) are given concentrations of copper, gold and silver in samples from the surface and particular intervals within the drill holes.

Table 1. *Concentration of copper, gold and silver in particular samples from the Borović locality*

Sample	Cu (%)	Ag (ppm)	Au (ppm)
TS-1	0.032	0.65	0.10
TS-2	0.062	1.20	0.19
TS-3	0.024	1.05	0.45
TS-4	0.030	0.95	0.43
KB-1	0.042	0.60	0.17
KB-2	0.040	0.90	0.62
KB-3	0.044	0.95	0.18
KB-4	0.078	0.85	0.20
KB-5	0.028	0.85	0.10
KB-6	0.028	0.90	0.10

As it can be seen from the table above, the mineralized area points out to a porphyry system characterized by low concentrations of representative metals. We have to mention that lower concentrations of copper are uncommon, since locally they are significantly higher and microscopic studies (of lower intensity too) have been determined copper mineralizations represented by presence of chalcopyrite. Also, at surface on wider areas have been determined so-called copper blossoming represented by Cu-carbonate minerals such are malchite and azurite. There are indications that the most of copper at this shallow and surface levels was washed out (Klajn, 1977). So, once again we stress out, that the majority of these samples were taken from the surface or shallow drill holes and probably should be expected that to depth the copper concentrations will rise.

Silver concentrations that have been within the range of 1 ppm are completely compatible with those usually found in porphyry copper deposits worldwide. Gold concentrations are very similar to those found at Plavica and Crni Vrv localities, while in particular samples are very indicative (TS-3 and KB-2). Without any doubts such gold concentrations are pointing out to an existence of favorable environments for disposal of epithermal gold mineralizations mainly relate to the zones of faulting and silicification. This should be used as a guidline for planning and directing further detailed exploration in this area.

In the close vicinity of the Zdravevci-Borović system itself, the oldest and deepest rocks are Paleozoic and Precambrian magmatic and metamorphic rocks (schists and amphibolitic rocks) of Serbo-Macedonian mass, and granodioritic pluton bodies (Klajn, 1977; Rakić et al., 1995; Rakić et al., 1993). Estimated temperature in this source is about 200 ºC. These rocks are covered by Eocene and Oligocene sedimentary, volcanic and sedimentary-volcanic rocks, mostly clastic rocks and flysch (Figure 1), plus volcanic intrusions of dacite-andesite and diorite. Conglomerate and breccia prevail in Eocene and Middle Oligocene is represented by sandstones, breccias and limestones with high amount of volcanic material. Miocene is composed of various sandstones, marls and tuffs, Pliocene mostly by clastic rocks, and Quaternary by alluvium and delluvium. All these rocks are dissected by andesitic and dacitic eruptive rocks, tuffs and ignimbrites. Complete thickness of rocks above the pluton is about 1000-2000 m (Mičevski et al., 2007).

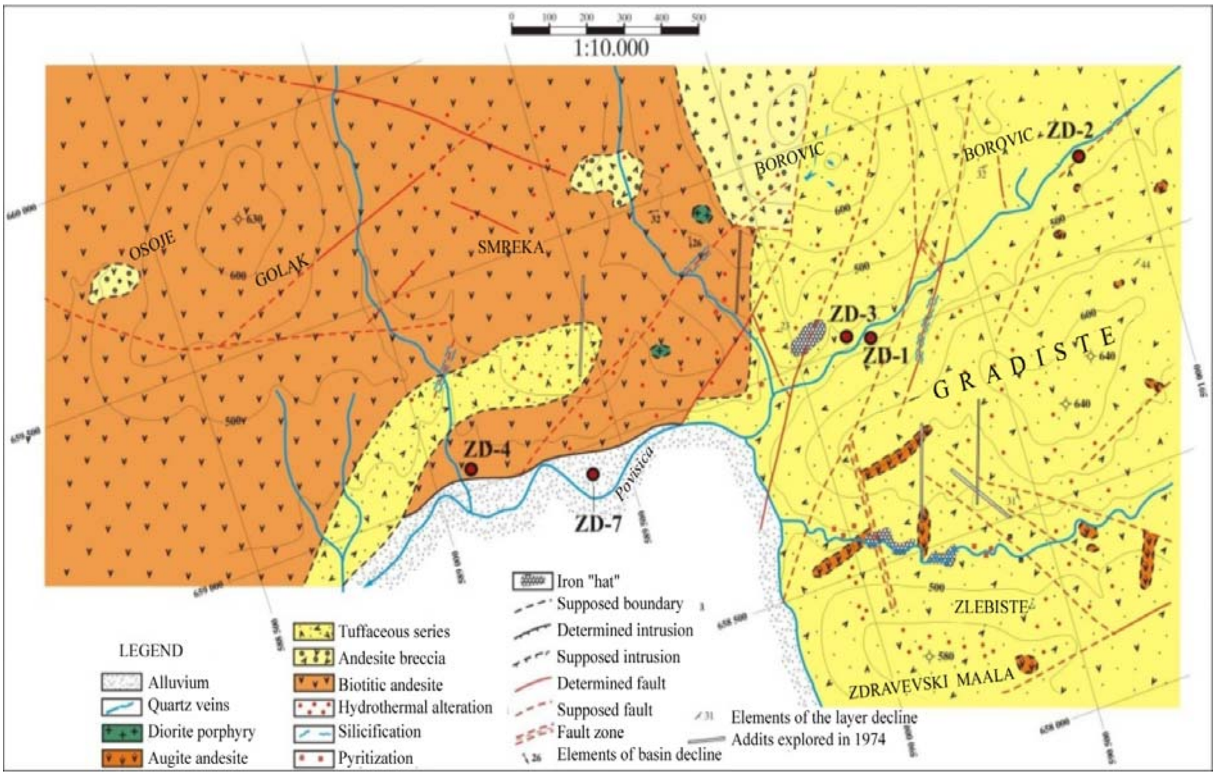

Figure 2. Simplified geological map of Zdravevci-Borović area with locations of analyzed wells (modified after Klajn, 1977).

Wells (figure 1) were drilled to various depths: well ZD-1 to 90 m, ZD-2: 107 m, ZD-3: 335 m, ZD-4: 134 m, ZD-5: 129 m, ZD-6: 96 m and later wells ZD-7 and ZD-9 (depths unknown). The major borehole ZD-3 lies 444 m above sea level in the valley of Povišnica and was drilled in 1989. Borehole diameter is 86 mm with installed casing of 3,5 inches. The well is drilled in a fracture zone and is relatively deeper than other surrounding boreholes.

Water was recognized early in 1991 as therapeutic (Milenković, 1991). Water is artesian and flows from the well ZD-3 with approximate discharge of Q = 10–11 l/s. The temperature of the water varies with depth and due to interaction with surface water and lies in the range of 43-48 ºC. Maximum temperature in the well was 50,8 ºC. Two aquifers have been found to exist in the area, first in the depth of 83 m and second on 210 m (Đuzelovski, 1999).

METHODS AND MATERIALS

Waters were sampled in two periods, first on 22.06.1991 and later in summer 2005. In the first period, only the major well ZD-3 was sampled, with intention to check the possibility of water for therapeutic use

(Milenković, 1991). Second sampling was done in 2005, and water samples from three wells ZD-3, ZD-7 and ZD-9 (Figure 2) were analyzed for geochemical composition and isotopes, plus the river Povišnica. Note that the results from the 2005 analysis for the well ZD-3 are not presented in this study, as sulphate concentration was far too low from expected and also far too low from the previous analysis in 1991, so this analysis was omitted.

Figure 3. Natural discharge of the well in Zdravevci-Borović area. Note the lack of vegetation in the background due to hydrothermal alteration and iron mineral precipitates in the creek bed in the foreground.

Analyses from the year 1991 were performed in laboratory in Belgrade, Serbia and analyses in 2005 in the ActLab laboratory in Canada. Measurements of water temperature and pH were determined in the field. Isotope composition of δD and $\delta^{18}O$ was performed in Ljubljana, Slovenia. As the concentration of HCO_3 ion (for the 2005 analyses) was not performed, it was calculated as the missing ion in geochemical software AquaChem via calculation of electrical balance to achieve electroneutrality. Saturation indices were calculated by software PHREEQC for Windows.

RESULTS AND DISCUSSION

Waters from wells belong to Ca-Na-HCO_3-SO_4, Na-Ca-HCO_3-SO_4 and Ca-HCO_3-SO_4 types and therefore their origin can be attributed to carbonates-sulphates with elevated Na values from volcanic complex, reflecting the groundwater composition.

First of all we have performed a statistical analysis of $\delta^{18}O$ and δD data, to see does the data follow certain standard and formerly determined rules. The data obtained for $\delta^{18}O$ and δD in water samples were plotted on a summary diagram of isotope compositions of waters of different origins (see Taylor 1967; Brownlow, 1996 and Misra, 2000) with a primary goal to determine the exact origin of waters of interest (Figure 3). As can be seen from plot bellow (Figure 3), there were slight discrepancies from the MWL (Meteoric Water Line) determined by the equation $\delta D=8 \, x \, \delta^{18}O +10$, but however the grouping of data was around the MWL. Those dicrepancies reflect the direct fractionation of meteoric water of O and H in contact with adjacent rocks and deep hydrothermal fluxes.

The plot have shown that majority of data plots are on or near the MWL indicating the meteoric origin of waters for most of the samples. Few of the samples, as can be seen from the diagram, have shown that there is a direct interaction and isotopic exchange between the meteoric generated waters and hydrothermal systems. This was direct confirmation about the nature of fluids, which have played the major role in the mineralization processes that have generated the polymetallic deposit. The main conclusion from this part can be drawn that there was an interaction between the meteoric water and a certain magmatic intrusion, which acted as an heat engine that have initiated an hydrothermal convection in the groundwater of the enclosing country rocks.

From the isotope data it is clear that although deviations from the Meteoric Water Line (MWL) $\delta D = 8 \text{ x } \delta^{18}O + 10$ occur (Figure 4a and b), samples are still grouped along this line. Water is therefore believed to be infiltrated meteoric water, which infiltrates through several fractures in sedimentary and volcanic rocks, is progressively heated in the depth and is later captured in the wells near the surface.

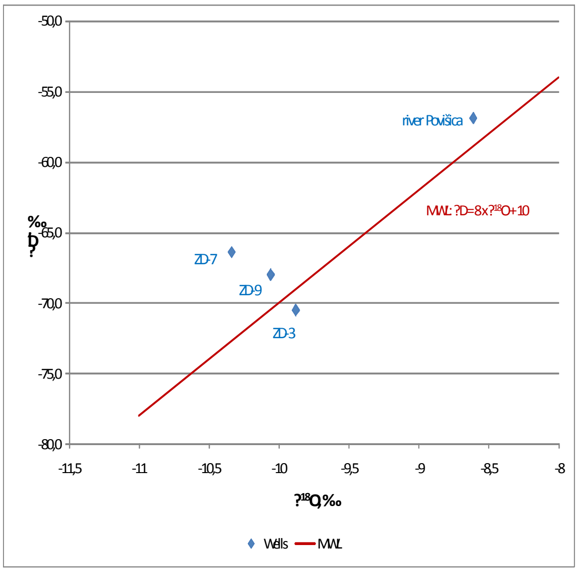

Figure 4a. Isotope composition of Zdravevci-Borović wells ($\delta^{18}O$ and δD).MWL: Meteoric Water Line.

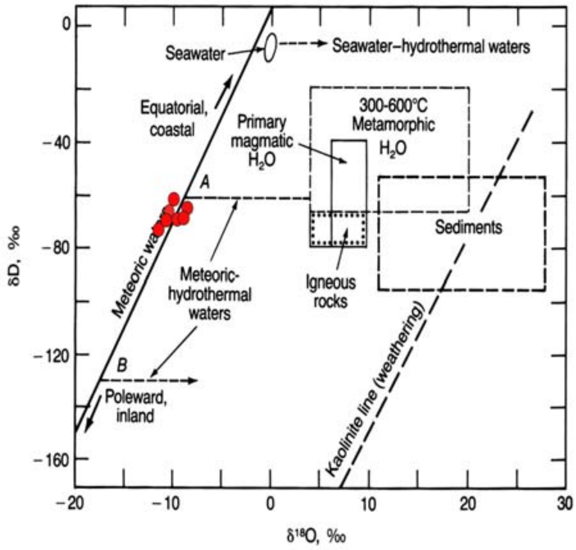

Figure 4b. Diagram of isotope composition of waters of different origins with included plots of values obtained for Borović samples (Taylor, 1974; 1997)

Water is highly mineralized, as the total dissolved solids value (TDS) is 3458 mg/l (based on the 1991 analysis). Water composition is presented on the Piper and Schoeller plots (Figure 5), and water from all wells belongs to one group.

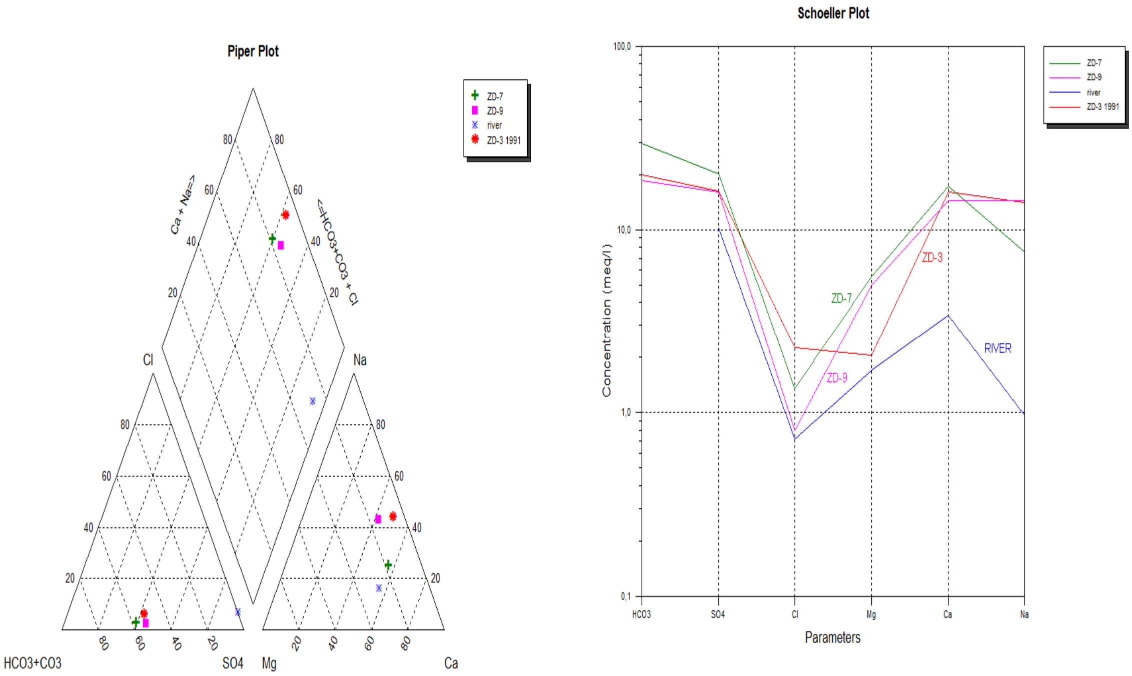

Figure 5. Piper and Schoeller plots of hydrogeochemical analyses of Zdravevci-Borović waters.

Temperature from deepest well ZD-3 was 43-44 ºC, but significantly lower in other two wells (21,2 ºC in well ZD-7 and 22,8 ºC in ZD-9). The reason for relatively cooler water can be attributed to lower well depth of two wells, as infiltrated water is progressively heated in greater depths. Deeper well ZD-3 is also situated in a more fractured zone, which permits higher circulation rate of water. Obviously, the potential for geothermal exploitation of the area exists, as the water outflows naturally from the wells (Figure 3). If pumped, the cooling effects would be less pronounced and temperature would be higher on the surface.

Table 2. *Selected concentration of water wells and river Povišnica and saturation indices of calcite, gypsum, goethite and hematite. Bold numbers indicate exceeded drinking water limit values.*

Well	pH	Na	Fe	Mn	Ni	As	Cu	Pb	Zn	Br	F	SO4	SI_{cal}	SI_{gyp}	SI_{goe}	SI_{hem}
unit	-	*mg/l*	*mg/l*	*µg/l*	*µg/l*	*µg/l*	*µg/l*	*µg/l*	*µg/l*	*µg/l*	*mg/l*	*mg/l*	-	-	-	-
river	**5,7**	22,4	0,19	**50,1**	5,7	1,73	**3,7**	2,25	24,1	26	0,3	**488**	-	-	-	-
ZD-3	6,5	**324,5**	**0,90**	**300**	5	**40**	**5**	5	30	0	**1,8**	**780**		-		
1991													0,52	0,53	6,97	16,02
	5,8	173	**5,15**	**872**	**25,1**	**49,6**	**4,0**	0,72	9,8	**131**	**1,8**	**971**	-	-		
ZD-7													0,29	0,44	4,37	10,72
	6,0	**333**	**3,93**	**350**	**69,2**	**60,6**	**8,4**	0,43	7,7	**145**	0,8	**767**	-	-		
ZD-9													0,30	0,56	5,05	12,11
limit	*6,5-9,5*	*200*	*0,2*	*50*	*20*	*10*	*2*	*10*	*-*	*10*	*1,5*	*250*	-	-	-	-

Despite the location in the regional area of highly mineralized ore deposits with Pb-Zn and Cu, and the fact that such surface waters can be highly contaminated with metals like Cu and Zn values for the the waters in Kratovo-Zletovo area (Alderton et al., 2005), the analyzed waters do not show exceeded values of permissible drinking water element concentrations for Pb or Zn. However, several other metals and parameters are exceeded (Table 2) in regard to EU Drinking Water Directive (98/83/EC). Values of Fe, Mn and SO_4 are greatly exceeded in all wells, and some other parameters also exceed the permissible limits for Cu, Na, Ni, As, Cu and F. The pH values are also a bit lower than lower limit. Selected calculated saturation indices (Table 1) indicate that the water is greatly oversaturated by iron oxides and hydroxides (SI>10), and slightly undersaturated or in relative equilibrium with calcite and gypsum (SI is around ±0,5). Iron minerals are therefore expected to precipitate from the water, and such precipitates are indeed visible in the field (Figure 3).

CONCLUSIONS

The Borovic locality represents one of the most potential areas within the Kratovo-Zletovo volcanic area from the aspect of Cu-Au mineralizations and geothermal-thermomineral waters.

Intensively hydrothermaly altered rocks, manfested by silicification, kaolinitization, sericitization, limonitization, etc., are important indicator of ore mineralization and existence of the geothermal system. Intensively silicified volcanic tuffs are characterized by gold contents of up to 0.46 g/t Au.

The investigated area of Zdravevci-Borović geothermal system is potentially interesting for geothermal water exploration and exploitation, as water from wells has temperatures up to about 50ºC, with discharge of approximately 10 l/s. With the use of submersible pumps, even higher temperatures at higher discharges can be obtained.

Origin of water can be related to sedimentary (carbonate) and volcanic cover due to its geochemical composition, and the water is attributed to be meteoric as it was shown by isotopes of hydrogen and oxygen or their respective ratios H/D and $\delta^{18}O$, which values were within the range -10.62÷ -8.61 $\delta^{18}O$ and -72.70÷ -56.90 δD. These values of studied isotopes are compatible with hydrothermal fluid systems, which have produced mineralizations at the Borović area.

Water infiltrates in fractured rocks, is progressively heated in the depth and captured at the surface at relatively high temperatures (highest in well ZD-3).

By further isotope analyses (tritium), the age of water and cycling could be determined. Systematic geochemical sampling of water from wells should be performed in the future to get further information on the water-rock interaction, to study the speciation and mobility of elements and evolution of ground water. The wells should be hydraulically tested for influence to determine the effects of pumping on temperature and discharge.

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