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PHYSICO-CHEMICAL CHARACTERISTICS OF GEOTHERMAL WATER IN TULARE NEAR MEDVEĐA

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Abstract. *The paper presents general characteristics and legal regulations for the use of thermomineral waters in Serbia, provides information on the regional position and geological characteristics of Tulare near Medveđa, the springs of water in Tulare and the methods of testing the physical and chemical characteristics of water in Tulare are described. In addition, the results of measuring the temperature, hardness, acidity, and chemical composition of water in Tulare are given. The results of the measurements showed that the water in Tulare belongs to homeothermal waters, that is soft to very soft, slightly acidic, and has an increased amount of Na, Ca, K and Mg, and also Sr, B, Li, P, As and Hg in small quantities. The water from the spring in Tulare is not suitable for drinking. To determine the balneological utility of the water in Tulare it is necessary to carry out appropriate drilling and further examine the physical, chemical, and balneological properties of this water.*

Key words: *Tulare, thermo-mineral waters, spas, minerals*

1. INTRODUCTION

The use of thermomineral (TM) waters in Serbia has a far longer history than its scientific research. Material remains and preserved cultural heritage from the Middle Ages are located near TM sources (monasteries, old towns). During the Ottoman rule from the 15th to the 19th century, bathrooms, known as hammams were built around thermo-mineral springs in Serbia (Protić, 1995).

Geologists, hydrogeologists, geographers, climatologists, chemists, doctors, balneologists, tourism masters, and others have produced a large number of written materials about our spas. Literature about spas, i.e. mineral, thermal, and thermomineral springs, originates from different periods, scientific - professional specialties, and application procedures in the field.

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Spas in Serbia represent its significant tourist value, all the more important since the health factor for many is still the main motive for the spa visit. The role and significance of the spas have changed significantly compared to past times. Instead of typical health centers, spas become places for rest and recovery of a wide range of people, seeking an ecologically healthy environment and a beautifully decorated ambiance (Stanković, 2011).

Scientific papers from the first decade of the 21st century deal with TM springs in Serbia from the aspect of their use as a renewable energy source, the possibilities of substitution of fossil fuels, the radioactivity of water, exploitation in spa tourism, conditions and possibilities of using thermo-mineral waters for balneological purposes, etc. (Milivojević et al., 2005; Joksimović et al., 2013).

The most important tourist resorts in Serbia have developed around thermal mineral resources in the river basin of the Western Morava and South Morava. These are typical European spas located next to the sources of thermomineral waters in the suburban zone. In the reservoirs, rehabilitation centers and specialized hospitals with thermomineral water are built. However, the tourist function of the spa is secondary due to the usual traditional spa concept in Serbia (Marković, 1980; Jovičić, 2008).

Thermal springs in the South Morava basin, which have been affirmed as spa and tourist sites, are Vranjska banja, Sijarinska banja, and Prolom banja. Stanković, in his research, provided ample information about the water springs in Tulare, which are related to the Radanski andesitic complex, which also belong to the springs in Sijarinska, Vranjska, and Prolom banja. There is still no scientific and professional literature on thermomineral springs in Tulare (Stanković, 2009).

1.1. General characteristics of the thermomineral waters

Mineral water is healing water that contains in one liter more than one gram of dissolved mineral matter. Thermal waters include a group of mineral waters that differ from their "ordinary", cold water by their temperature. Thermomineral waters are waters of temperatures higher than 20 °C, which are divided into hypothermal (20-34 °C), homeothermal (34-38 °C), and hyper-thermal (over 38 °C) (Dukić, 2008).

SI water hardness units are expressed in $mgCaCO_3/dm^3$ water. However, in our country hardness of water is expressed in German degrees °dH. Depending on the degree of hardness, waters can be very soft (0-5 °dH; 0-70 $mgCaCO_3/l$), soft (5-10 °dH; 70-140 $mgCaCO_3/l$), moderately hard (10-15 °dH; 140-210 $mgCaCO_3/l$), hard (15-25 °dH; 210-320 $mgCaCO_3/l$) and very hard (>25 °dH; 320-540 $mgCaCO_3/l$) (Velagić, 1997; LaMoreaux, 2001; Petrović, 2010; Petrović, 2012).

According to pH value waters are divided into very acidic (<3.5), acidic (3.5-5.5), weakly acidic (5.6-6.8), neutral (6.8-7.2), weak alkaline (7.2-8.5), and alkaline (> 8.5).

1.2. Legal regulations

In Serbia, the Ordinance on the hygienic correctness of drinking water (Official Gazette of FRY no. 42/98 and 44/99), which refers to water intended for human use, is in force. According to this Ordinance, hygienically correct drinking water is the water with the maximum allowable values of As (0.01 mg/l), Hg (0.001 mg/l), and Pb (0.01mg/l) (RHDWQ, 1999).

The Law on Spas of the Republic of Serbia (Official Gazette of the Republic of Serbia, no. 80/92 and 67/93) prescribes that a spa is an area in which it is located and one or more natural healing factors is used and which meets the requirements regarding the regulation and equipment for their use. Following natural healing factors are considered: thermal and mineral water, air, gas, and medicinal mud (peloid), whose medicinal properties are scientifically tested and proven in accordance with this Law.

1.3. Brief overview of individual thermomineral spas in Serbia

1.3.1. Vranjska banja

Vranjska banja (Vranjska spa) is located at 42°33'11" north latitude and 21°59'19" east longitude, in a spacious Vranje basin in southeastern Serbia at 420 meters above sea level, 10 kilometers east of Vranje. According to the political-administrative division, Vranjska banja is located within the Pcinja district, which is the seat of the city of Vranje.

Hot springs of Vranjska banja are the warmest in Serbia whose water temperature ranges from 63 °C to 95 °C, and the water vapor temperature reaches 110 °C. According to this, Vranjska banja is one of the warmest spas of Europe with hyperthermal water. So far, 10 springs have been captured, out of which 4 are larger springs of geothermal water of different temperatures and compositions, with a yield of 6-44 l/s. The total volume of the springs in Vranjska banja A-1, A-2, VG-2, and VG-3, according to the previous tests, is up to 60 l/s. The geothermal water from the well VG-2 with a temperature of 95 °C and a yield of up to 27 l/s was examined. The springs B-1, A-3, B-2, *Gornji izvor*, and *Sabirni kanal* have slightly lower average yields, around 20 l/s. *Gvožđeviti izvor*, *Izvor u potoku*, *Gornji*, and *Glavni izvor* have a water temperature ranging from 61 to 86 ºC.

1.3.2. Jošanička banja

Jošanička banja (Jošanička spa) is located at 43°23'21" north latitude and 20°45'08" east longitude at the foot of the mountain Kopaonik, at 550 m above sea level, in the valley of the Jošanica River and its tributaries Samokovka. According to the political and administrative division, Jošanička banja belongs to the Raska district, which is the seat of the municipality of Raska 25 km away. It is 30 km from the peak of Kopaonik, which is why it is often called the *Green Gate of Kopaonik*. Jošanička banja is in the protected area (Banjski borjak) within the National Park Kopaonik.

Geothermal springs of mineral water in Jošanička banja are among the hottest in Serbia where according to water temperature they are only lagging behind Vranjska banja. The waters of Jošanička banja according to the temperature of water belong to the group of hyperthermal waters where three springs are separated. *Glavno izvorište* is in the village of Jošanička banja on the left side of the Jošanica River with 11 springs. *Drugo izvorište*, of the lower capacity, is on the right side of the river Jošanica and consists of 3 springs and third, *Slanište*, is located at a distance of 1850 m from the center of the spa, downstream by the river Jošanica.

The warmest is *Glavni izvor* with a water temperature of 78.5 °C and a yield of 7 l/s. Other springs obtain different water yields and temperatures ranging from 36 °C to 75 °C. The spring on the right side of the Jošanica River has three springs of poorer yield, a total of about 3.1 l/s, and a temperature of up to 70 °C. The salinity is made up of weaker springs of total yield up to 2.1 l/s and temperatures ranging from 28 to 37 °C. At 500 m upstream from the center of the Jošanička banja, there is *Luški potok*, a spring of cold iron-carbon water temperature of 14 °C, and a volume of 0.08 l/s.

1.3.3. Lukovska banja

Lukovska banja (Lukovska spa) is located at 43°58'67" north latitude and 21°27'83" of the eastern longitude within the village Lukovo, Kuršumlija municipality. Thermomineral springs of Lukovska banja are located on the eastern slopes of the mountain Kopaonik at an altitude of 681 meters.

According to the number of warm and mineral springs, Lukovska banja is the richest spring in Serbia. There are 37 hyperthermal mineral springs at Lukovska banja, temperatures from 22 °C to 65 °C, and they occur in a broken spring, in a narrow zone at a length of about 400 meters. In the period from 1977 to 1992, more detailed hydrogeological investigations were carried out in Lukovska banja with the drilling of 25 shallows (from 2 to 4 meters) and 6 deep (from 150 to 350 meters) wells, out of which 4 yielded 30 l/s of water whose temperature ranged from 59 to 63 °C.

1.3.4. Kuršumlijska banja

Kuršumlijska banja (Kuršumlijska spa) belongs to the municipality of Kuršumlija in Toplički district. It extends between 43°03'22" north latitude and 21°15'14" east longitude at 442 m above sea level. Kuršumlijska banja is located 11 km southwest of Kuršumlija.

According to the analysis of the Balneo-Climate Institute from Belgrade, performed at the end of the 20th century, there are ten mineral springs in Kuršumlijska banja, with a temperature of up to 62 °C and a yield of 11.1 l/s. Kursumlija banja waters belong according to their chemical composition to alkaline sulfur hyperthermal waters. It was found that Kuršumlijska banja water has interesting properties, especially due to its high content of silicic acid up to 111 mg/l, metaboronic acid up to 80 mg/l, lithium to 3 mg/l, fluoride up to 4.5 mg/l, hydrogen sulfate to 5.6 mg/l, carbonic acid 760 mg/l, etc.

1.3.5. Prolom banja

Prolom banja (Prolom spa) is located 23 km southeast of the municipality of Kuršumlija, which administratively belongs to Toplički district. It extends between 43°02'10" north latitude and 21°24'03" east longitude.

Prolom banja is located on the north-western slopes of the Radan Mountain at 550 to 668 m above sea level. Thermomineral waters appear at crossroads of the main fault whose main route is west-northwest with two side transverse faults, about 950 m apart. Prolom banja with temperatures from 25.5 °C to 38.5 °C belongs to a group of hypothermal waters. According to the degree of content of 0.32 g/l, this water is considered to be low mineralized.

1.3.6. Niška banja

Niška banja (Niška spa) is a municipality located in southeastern Serbia at 43°17'37" north latitude and 22°00'29" east longitude. It is 10 km from the center of Niš. It is located on the slopes of Koritnik Mountain at 250 m above sea level. It extends along the southeastern edge of the Niš valley. Of all spas in Serbia, Niška banja has the most favorable and busiest position. From the road leading to Bulgaria, the spa is one kilometer away and 250 kilometers from Belgrade. Niška banja and Niš are connected by an asphalt road with two lanes and railroad tracks.

There are five springs of thermal water in Niška banja: *Glavno vrelo*, *Suva banja*, *Školska česma*, *Pasjača* and *Banjica*. All springs belong to the group of earth-alkaline homeothermic temperatures of 36-38 °C, slightly mineralized, with a yield capacity of 56 l/s. The three main springs are: *Glavno vrelo*, *Suva banja*, *Školska česma*.

1.3.7. Vrnjačka banja

Vrnjačka banja (Vrnjačka spa) spans between 43°37'25" north latitude and 20°53'37" of the eastern longitude. It is located in the central part of Serbia, in the valley of the West Morava River, near the mountain Goč, at an altitude of 230 meters.

Vrnjačka banja has seven known mineral springs belonging to alkaline, carbonic acid homeothermal waters with a temperature of up to 36.5 °C and a capacity of 8.1 l/s. Four springs are used for balneological therapy: *Topla Voda*, *Snežnik*, *Slatina*, and *Jezero*, and the remaining three springs are for drinking only: *Beli Izvor*, *Borjak*, and *Vrnjačko Vrelo*. The spring *Snežnik* has a yield of 0.49 l/s and a temperature of 17 °C. The spring *Slatina* has a water yield of 1.14 l/s, with a temperature of 14 °C. Mineral waters of *Snežnik* and *Slatina* belong to the group of cold, alkaline, alkaline earth and coal-acid waters (Stanković, 2009).

1.4. Regional location and geological characteristics of the Tulare area

In the area of Serbia, the old tectonic masses of the Rhodope and the Pannonian shores and the younger spherical orogenic zone of the Dinarides and the Carpatho-balkanids are touching. At these contacts, in the fault zones, thermal and mineral sources appear. Therefore, a large number of thermo-mineral sources are related to the fault lines of the Rhodope masses in the area of southeastern Serbia, where the springs of Tulare are located as well (Rodić et al. 1994).

Tulare is at 42°37'21" north latitude and 21°27'19" east longitude at the foot of Mount Goljak.

Tulare is located in a small extension of the Tular River valley, the tributaries of Jablanica, at an altitude of 570 meters. Tulare is 10 km from Medveđa, 15 km from Sijarinska Spa, and 63 km from Leskovac (Figure 1). According to the 2011 population census, there were 162 inhabitants in Tulare (RIS, 2011).

Fig. 1 Physical – geographical location of Tulare

In the Tulare area, there are volcanic rocks of the Radenci andesitic complex, where already explored springs of Sijarinska banja belong. In the eastern part of Tulare, there are springs of warm mineral water, but until now the physical-chemical characteristics of these waters have not been studied (Stanković, 2009).

For the first time in Serbia, this paper investigates the physical and chemical characteristics of geothermal water from surface springs in Tulare near Medveđa.

2. Experimental Part

Investigation of temperature, hardness, pH, and mineral composition of water was carried out on Spring 1 and Spring 2 in Tulare.

2.1. Spring 1

Spring 1 is located in the western part of the village of Tulare, 350 meters from the main road Medveđa - Priština. The spring can be reached by the unpaved but passable way.

On the terrain, it was noticed that Spring 1 gashes water from 40 to 150 cm above the surface of the earth with interruptions of one minute (Figure 2). Spring 1 gives about 9000 liters of water in 24 hours.

Fig. 2 Water bursting in Spring 1 in Tulare

2.2. Spring 2

Spring 2 is located 50 meters from Spring 1. Water from Spring 2, with a yield of 6 l/s is pumped through a plastic tube into a collection pool (Figure 3)

Fig. 3 Spring 2 water flowing into a collection pool in Tulare

2.3. Measurement of water temperature

To measure the water temperature, a mercury-in-glass thermometer was used of the Dutch brand ROMED, with a measurement range of 32 ºC to 44 ºC, and the accuracy was ±0.01 °C.

2.4. Measurement of water hardness

The ECEMETAR brand ADWA was used to measure water hardness, type AD203 battery type 150 Hrs/4x1.5V, at an operating temperature from 0 to 50 °C and relative humidity of 95%. Measurement accuracy is ±0.02 °dH.

2.5. Water pH measurement

To measure the pH of water, a digital pH meter ME-PH006 was used, with a measurement range of 0 to 14 pH, at a working temperature of 0 °C to 50 °C. Measurement accuracy is ±0.03 pH.

2.6. Determination of macro, micro, and toxic elements

To determine the mineral water composition, an Inductively Coupled Plasma-Optical Emission Spectrometer-ARCOS FHE12, SPECTRO, Germany (ICP-OES, 1.4 kW, 12 MHz) was used. The ICP-OES analysis was performed by measuring the intensity of radiation of the specific wavelengths emitted by the elements. The samples were dispersed in a liquid and introduced into the plasma in the form of an aerosol, where they were vaporized, atomized, and excited. The carrier gas was Argon 5.0 (99.999% purity). The total uncertainty (including accuracy error) of the analysis is less than 2%. To estimate the content of all examined elements, calibration standards were prepared. Multistandard, a standard solution (Merck) containing Ag, B, Ba, Bi, Ca, Cd, Co, Cr, Cu, Fe, K, Li, In, Mg, Mn, Na, Ni, Pb, Tl, and Zn at a concentration of 1000 ppm, was used for the preparation of calibration solutions. The elements As, Hg, P, and Sr were determined using specific calibration standards (1000 ppm), respectively. Distilled water, purified by Fisher Chemical (HPLC grade), was used for all sample dilutions. The preparation of standard solutions was performed by diluting the multistandard so that the concentrations of the standards for the calibration were in the range of the expected concentrations of the analyzed elements.

3. Results and Discussion

The results of measuring the temperature, hardness, and pH of the water samples from Spring 1 and Spring 2 in Tulare are given in Table 1.

Based on the data in Table 1, water from Tulare belongs to the group of homeothermal waters. In terms of temperature, water in Tulare can be used in agriculture and greenhouses, fish farming, spas and bath facilities, district water heating, soil warming, fruit and vegetable drying, concrete curing, food processing (Dokmanović, et al. 2012).

Based on water hardness data in Table 1, the water from Spring 1 in Tulare is located at the border between soft and very soft water. Water from Spring 2 belongs to soft water. The hardness of the water from the Springs in Tulare comes from dissolved salts of calcium and magnesium in water.

Table 1 The results of measuring the temperature, hardness, and pH of the water samples from Spring 1 and Spring 2 in Tulare

Water samples	Water temperature (°C)	Water hardness (°dH)	pH value
Spring 1	35	4.6 - 5.9	5.7
Spring 2	37.5	6.4	5.9

Based on the data for the pH in Table 1, the waters from Spring 1 and Spring 2 in Tulare belong to slightly acidic waters.

The results of determining the chemical composition of water samples from Spring 1 and Spring 2 in Tulare are given in Table 2.

Based on the data in Table 2, it can be seen that the waters from the Spring 1 and the Spring 2 in Tulare are rich in macroelements (Na, Ca, K, Mg) and that there are Sr, B, Li, and P microelements, while Bi, In, and Zn occur in traces.

Sodium is suitable for the production of hydrochloric acid in the stomach, the treatment of skin and chronic kidney disease, etc. Calcium plays an important role in the operation of the heart muscle and the regulation of the level of blood sugar and iron. Potassium is important for bone mineralization and muscle activity. Magnesium has a calming effect on the central nervous system and has a positive role in the treatment of the urinary tract and the secretion of enzymes, for the proper functioning of the pancreas and liver, etc. Magnesium and calcium positively influence the increase in the number and mobility of spermatozoa. Microelements Sr, B, Li have a positive effect in the treatment of stomach and digestive tract diseases (Štraser 1953; Stanković, 1983; Jovanović, et al. 1994; Milićević, 1987; Rakić, 1987; Maćejka and Tanasković, 1994; Protić, 1995; Jovanović et al. 2001; Komatina, 2001; Mihajlović, 2002; Dragović, 2007).

Table 2 Results of determining the chemical composition of water samples from Spring 1 and Spring 2 in Tulare

Chemical element	Spring 1 water mg/l	Spring 2 water mg/l
As	0.031	0.041
Ag	not detected	not detected
B	17.122	13.417
Ba	0.066	0.036
Bi	0.049	0.045
Ca	191.620	225.801
Cd	not detected	not detected
Co	not detected	not detected
Cr	not detected	not detected
Cu	not detected	0.001
Fe	0.534	not detected
In	0.018	0.025
K	71.965	46.384
Li	2.692	2.166
Mg	29.693	31.956
Mn	0.007	0.253
Na	83470.411	55518.200
Ni	not detected	not detected
Pb	not detected	0.004
Sr	18.538	13.921
Tl	not detected	not detected
Zn	0.028	0.022
Hg	0.044	0.039
P	0.732	0.766

It is interesting to note that iron is detected only in water from Spring 1, and magnesium in water from Spring 2 in Tulare.

Based on the data in Table 2, it can be seen that the concentration of As (0.031 mg/l) and Hg (0.044 mg/l) significantly exceeds the maximum permitted concentrations provided by the Regulation on hygienic correctness of drinking water (RHDWQ, 1999). In this regard, the water from the Spring 1 and Spring 2 in Tulare should not be used for drinking without proper purification from the mentioned metals.

A comparative overview of temperature, hardness, and pH of water from Spring 1 and Spring 2 in Tulare and geyser in Sijarinska banja is shown in Table 3.

Water from the geyser in Sijarinska banja originates from a well at a depth of 1232 meters. It should be noted that at a depth of 300 m, the flow rate of 60 l/s appeared. For further examinations, the boring went deeper and reached the depth of 1232 m. At this depth, the flow rate was about 33 l/s, and for that reason, it was damped (Stojiljković, 2006; Stojiljković, et al. 2015).

Based on the data in Table 3, it can be seen that the water temperature in Sijarinska banja is significantly higher than the water temperature in Tulare and that the water in Sijarinska banja is harder than water in Tulare. It can also be seen that the water from the Spring 1 and the Spring 2 in Tulare is slightly acidic, and the water from the Sijarinska banja is poorly alkaline.

A comparative overview of macroelements from Spring 1 and Spring 2 in Tulare and geyser in Sijarinska banja is given in Table 4.

Based on the data in Table 4 it can be seen that the concentrations of macroelements (Na, Ca, K, Mg, Sr, B, and Li) from the Spring 1 and Spring 2 in Tulare are significantly higher than the concentration of the same elements in the water from the Sijarinska banja. The concentration of Na, Ca, K, and Mg in the water in Tulare is 72.2, 48.0, 27.7, 32.6 times higher than the concentrations of the same elements in water from Sijarinska banja.

Table 3 Comparative overview of temperature, hardness, and pH of water from Spring 1 and Spring 2 in Tulare and geyser in Sijarinska banja

	Spring 1 water	Spring 2 water	Sijarinska banja
Temperature (°C)	35	37.5	75-78
Water hardness (°dH)	4.6 – 5.9	6.4	9.24
pH value	5.7	5.9	7.5

Table 4 Comparative overview of macroelements from Spring 1 and Spring 2 in Tulare and geyser in Sijarinska banja

Chemical element	Spring 1 water mg/l	Spring 2 water mg/l	Water from Sijarinska banja mg/l
Na	83470.411	55518.200	1155.790
Ca	191.620	225.801	6.910
K	71.965	46.384	85.911
Mg	29.693	31.95	15.245
Sr	18.538	13.921	0.402
B	17.122	13.417	8.760
Li	2.692	2.166	1.957

For the balneological application of the water in Tulare it is necessary to conduct water testing from deeper layers of soil, perform its physical, chemical, and biological analysis, etc.

4. Conclusion

In the light of all the above said, it can be concluded that the water from the Tulare springs with a temperature of 35 to 37.5 °C belongs to homeothermal waters, with a hardness of 4.6-6.4 °dH it is at the limit of soft and very soft water, that with pH values from 5.7 to 5.9 it belongs to slightly acidic water and that there are Na, Ca, K, Mg macroelements, and Sr, B, Li, and P microelements. The concentration of Na, Ca, K, and Mg in the water in Tulare is significantly higher than the concentration of the same elements in water from the nearby Sijarinska banja.

Bearing in mind that the concentrations of As and Hg are significantly higher than the concentrations of As and Hg permitted by the Regulation on hygienic correctness of drinking water, it can be concluded that the water from springs in Tulare is not suitable for drinking.

In the future, drilling of the soil in Tulare should be carried out to the depths as in Sijarinska banja (1232 m), and the physical, chemical and biological characteristics of the water in Tulare from the wells should be performed.

Spas in Serbia are a significant tourist value, as one of the main factors of health and for many, this is the basic motive for visiting spas. It may be that further water tests in Tulare could show that in the future Tulare could be used as a spa resort and a new tourist attraction in Serbia.

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FIZIČKO-HEMIJSKE KARAKTERISTIKE GEOTERMALNIH VODA U TULARU U BLIZINI MEDVEĐE

U ovom radu su predstavljene generalne karakteristike termomineralnih voda kao i regulative za njihovu upotrebu u Srbiji. Opisan je regionalni položaj i geološke karakteristike Tulara, mesta blizu Medveđe, i izvora vode u ovom mestu; kao i metode za određivanje fizičkih i hemijskih karakteristika voda sa ovih izvora. Prikazan je hemijski sastav i rezultati merenja temperature, tvrdoće i kiselosti ovih voda. Rezultati merenja pokazuju da voda u Tularu pripada homeotermalnim vodama, koje su meke do veoma meke, blago kisele sa povećanom količinom Na, Ca, K i Mg, i takođe sadrže Sr, B, Li, P, As i Hg u malim količinama. Voda sa izvora Tulare nije za piće. Da bi se utvrdile njene balneološke mogućnosti neophodno je izvršiti odgovarajuća bušenja i dalje ispitati njihova fizička, hemijska i balneološka svojstva.

Ključne reči: *Tulare, termo-mineralne vode, banje, minerali*