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CHARACTERISTICS AND ORIGIN OF THE SPLIT SULPHUR SPA (SOUTHERN CROATIA)

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CHARAKTERISTIKA A PÔVOD SÍROVÝCH KÚPEĽOV V SPLITE (JUŽNÉ CHORVÁTSKO)

ABSTRACT

The objective of this paper is to study the hydrological characteristics and origin of the sulphur water from the Split spa in Southern Croatia in 1987, 1988 and 2003.

This paper presents the results of monitoring the content of chlorides and hydrogen sulphide as well as the sulphur water temperature.

Since the hydrogen sulphide content during the dry periods significantly differs from the hydrogen sulphide water content during rainy periods this paper also compares the results obtained for those two periods.

Under the influence of great quantities of rainfall during cold periods (winter and the beginning of spring) the ratio between seawater and surface water changes and thus the chlorides content and the content of other minerals are reduced and the lowered temperature reduces hydrogen sulphide which can disappear completely.

The concentration of 12‰ chlorides (76–94. 4% days/year) and 12 mg/L hydrogen sulphide (66,7 – 88. 9% days /year) has been taken as a limit value between water with a normal typical content and water in rainy, cold periods.

According to the monitoring results it can be concluded that sulphur water consists of seawater and hydrogencarbonate surface water with a fairly constant water content during dry periods while the hydrogencarbonate water content increases during rainy periods.

Key words: *sulphur water, hydrogen sulphide, Split spa*

1. Introduction

Mineral water is natural groundwater which differs from other types of groundwater according to the quantity or type of dissolved salts, temperature and radioactive matter content. This water has a higher content of some substances with strong physiological effects. Mineral water is generally classified as therapeutic water. Its therapeutic effect has to be scientifically proved after which it can be used as drinking water or water for bathing, inhalation, compresses, etc. Considering the purpose of the water use, its hygienic and sanitary safeness has to be ensured, i.e. its quality should correspond to drinking water quality. Mineral water originates from the natural hydrological cycle with slightly different physical and chemical characteristics and with a specific gas and mineral content. Consequently, this water originates from rainfall and later flows by gravity in the form of surface water or groundwater. Its gradual mineralization, increased by juvenile gases (Xu et. al., 1998) from a considerable depth, results from large watered contact surfaces and from its exceptional solubility as it flows through underground geological layers.

Thermomineral water is of atmospheric origin and after passing through rocks along faults, to a depth of 2500-3000 m where it is heated and enriched by minerals, it passes through younger faults and by ascending drains it comes to the surface and flows into the secondary aquifer (Slišković, 1998; Masakatsu & Fraser, 1995).

Thermomineral water is balneologically characterized by a higher mineral content, more than 1000 mg/L, and a temperature above 20^{o}C. Mineral water can be classified as alkaline, ferrous, chloride, carbonate, sulphate, sulphur water, etc. and can contain a high quantity of sodium chloride. If it contains more than 240 moles NaCl it is referred to as salty water (Levačić, 1997).
Water with more magnesium sulphates is referred to as bitter water (Levačić, 1997).

The therapeutic effect of mineral water has been recognized since ancient times. The biological effect of mineral water is important for the circulation of minerals since their presence in the human body is equally important as proteins, fats and vitamins. Although attempts have been made to produce artificial mineral water with the same content as natural mineral water it has not been possible to achieve the same therapeutic effect.

According to the dissolved mineral content or other ingredients this water contributes to the curing of specific illnesses (Leibetseder et al., 2004). The discipline dealing with this type of medical cure is defined as balneology. It is most frequently used to cure rheumatic illnesses and illnesses of the respiratory or gastro-enterologic tracts as well as many others.

It is generally known that even the least quantity of radioactive matter introduced into the human body, either by ingestion or inhalation, can cause harmful effects (WHO, 2001). This becomes particularly important in water with an increased content of radioactive substances since the use of such water, if not strictly controled, can have negative effects upon our health (ICRP, 1990).

Apart from the use of mineral water for drinking it can be used for compresses and baths, therefore, some types of mineral water are important worldwide and can influence the economic development of a country (Buick et al., 2002). In addition to the Split spa other spas in Croatia are located in Istria, Varaždin and Tuhelj, which contain ca 7 mg/L of hydrogen sulphide. The objective of this study dealing with the Split sulphur spring, was to analyze the content of the spring in order to use it for epidemiological comparative studies on the efficiency of the use of balneo-therapeutic spring water.

The mineral water from the Split spa is classified among waters with an increased concentration of hydrogen sulphide (at least 12 mg/L) (Drew, 1996). It is a mineral, sulphur, hypothermic, Na-Cl type of water. Numerous researchers believe that the Emperor Diocletian used the sulphur spring and that it was one reason for building the Palace at that site. This cannot be proved since there are no written documents. It is well-known, however, that the Romans used warm mineral springs for medical treatment (Ivanković, 1981). According to an international classification (Novak, 1968, 1969) the Split sulphur water is classified among the most highly-mineralized sulphur hypothermic types of water which contain at least 1 mg/L of hydrogen sulphide.

2. Study area

The Split spa is located near the sea shore in the city center ca 200 meters west of Diocletian's Palace. There are several springs and the main spring, Kupališno vrelo, is located in the south-western part of the city's fish-market which is ca 10 meters from the spa. The intake for the spring water is a well 2. 2 m deep. The water from that well is used for the Split Medical Center (Figure 1). The second spring, Samostansko vrelo, is in the cellar of St. Francis Monastery, about 300 meters of the spa and nearer the sea shore.

3. Methods

The chlorides and hydrogen sulphide in the sulphur water in the Split spa were monitored six times a month during 1987, 1988 and 2003. The quality parameters (temperature, chloride, sulphate, calcium, magnesium, hydrogencarbonate and hydrogen sulphide) in the samples were determined according to American Standard Methods (AWWA, 1975, 1995).

4. Results

The results (Table 1) were presented as mean concentrations ($\overline{C}$), standard deviation (σ), variation coefficients (CV,%) and ranges. Temperature, hydrogencarbonate, chloride, sulphate, calcium, magnesium and hydrogen sulphide were presented in the water from the spring intake structure and in the water from the water flowing channel (Table 2).

Table 1 also includes the statistically analyzed results obtained by the Institute for Oceanography and Fishing (1952/53) (Buljan, 1962).

The statistical analyses were performed taking into account the presence of two characteristically, different water types, i.e. water with a normal content for the period without long-lasting storms and water with a non-typical changeable content in the periods of abundant rainfall. As a limit value for those two types it was assumed that the normal water with a typical content has at least 12 mg/L H_2S, while the water with a non-typical content during heavy rainfall contains fewer chlorides and hydrogen sulphide (Štambuk-Giljanović, 1998). The results of the water analysis in the water from the spring intake structure and in stagnant water in the overflow channel during short droughts are presented in Table 2. It can be concluded that the water from the intake structure is a typical hydrogencarbonate type of karst water. The calcium hardness is fairly higher than magnesium hardness. This water contains more chlorides than typical karst water (Štambuk-Giljanović, 1998, 2005) which shows it is influenced by the sea. The sulphates content is slightly increased. The water from the overflow channel (Table 2) is diluted seawater since it contains an increased concentration of chlorides and more magnesium than calcium.

Figure 2 presents the individual results of the analysis of hydrogen sulphide and chlorides. The chlorides content (full line) is presented on the ordinate and the 10 day average rainfall is presented by a step diagram. The months during which the analyses was carried out are presented on the apscissa.

Table 3 presents the results of the temperature analyses for the three years. The highest temperature recorded at the Split spa sulphur spring was 23.75^{o} (30 September 1987) while the lowest was 14^{o}C (20 January 2004). During the entire period under study the water had hypothermic characteristics.

5. Discussion

The water from the Split spa had at least 12‰ chlorides (76 - 94.4% days / year) and 12 mg/L hydrogen sulphide (66.7 - 88.9% days / year) (Table 1). During the dry periods, the chlorides content ranged on the average from 16.5 to 17.7‰. There was no significant difference between the results obtained for the 1987, 1988 and 2003 period and those obtained for the 1952/ 53 period.

The results of the analysis of hydrogen sulphide and chlorides in the first half of the year oscillate more than in the second half, which is caused by the quantity of rainfall and temperature (Figure 2).

In the first half of the year, when the quantity of rainfall was greater and when temperatures were lower, the chlorides content decreased rapidly and hydrogen sulphide first decreased and then disappeared. The temperatures ranged from 14-16^{o}C. This occured in March 1987 and in February and March 1988 after heavy rainfall. It should be noted that in 2003 chlorides did not significantly decrease and hydrogen sulphide did not disappear which can be accounted for by a smaller quantity of rainfall than in previous years (Figure 2). The smallest chlorides content in 1987 and 2003 amounted to 1.2‰ while in 1988 the spring "dried up" for a short time. This phenomenon is quite frequent (Vierthaler, 1867; Glaser, 1909). The spring does not actually "dry up"; the groundwater inflow into the intake structure is decreased so that the water overflows through the overflow channel into the sea. This occurs during the lowest sea level after long dry periods, i.e. at the end of March when the spring inflow consists exclusively of surface water, ca 2-3 L/s, and the normal inflow is ca 15 L/s.

During this period when the spring "dries up" the water does not contain any hydrogen sulphide and the temperature is low. After the rise of the sea level, even with a small quantity of rainfall, the water inflow into the spring becomes normal so that the spring water contains a normal level of hydrogen sulphide or a slightly increased level (30 mg/L) up to 18000 mg/L chlorides (18‰) while the temperature reaches 23°C. The spring dries up. Baturić (1959) explained this phenomenom by low sea levels and accounted for the phenomenon of the normalization of the sulphur water content by its overflow from the underground basins under the influence of surface flows. The content of seawater and mineral water from the Split spa is similar (Table 4) and the water from the Split spa has a slightly higher content of calcium and sulphates. This results from some natural phenomena. However, both types of water differ considering the content of dissolved salts, i.e. the degree of mineralization. Seawater has ca 34 g/L, while mineral water from the Split spa has ca 28 g/L of dissolved salts and hydrogen sulphide. This shows that the mineral water from the Split spa contains both seawater and surface water.

Sulphur waters in Croatia (Table 5) differ according to their chemical content, degree of mineralization and according to hydrogen sulphide content. Since the mineral water spring of the Split spa is near the sea, the seawater influences its water.

The Varaždin water is, according to balneological characteristics, classified as mineral water, Ca-Na-HCO_3-SO_4, sulphur and hypertermic,while the Tuhelj water is classified as Ca-Mg-HCO_3, sulphur and hypothermic water (Novak, 1968, 1969).

According to Buljan (1962) sulphur spring water consists of seawater and mineral sulphate water which belongs to the type of water from the gypsum layers at Slana Jaruga in Glavica near Sinj. These two types mix with fossil organic matter such as bitumen, asphalt or lignite which are characteristic for the Eocen limestone region along the eastern Adriatic coastline. In this region the oxidation of organic matter (bitumen, asphalt) occurs under anaerobic conditions. Since the mentioned habitats have small quantities of air, oxidation occurs with the oxygen from the sulphates so that sulphate is reduced to H_2S.

Although the organic matter oxidation takes place at the point where seawater mixes with mineral surface water, Buljan states that this oxidation process is based on the sulphate from the mineral sulphate water. During the oxidation process some heat is liberated so that the spring water temperature increases. Buljan's conclusion (1962) about the existence of mineral sulphate water is based on the fact that the SO_4/Cl ratio in the sulphate water is higher than in the seawater in the Split port and Kaštela Bay. He also states that the calcium sulphate content is 14 - 62% higher in the sulphur water.

However, the investigation results presented in this paper show that the spring water, in the period when the spring dries up, i.e. during the lowest water level, belongs to the karst water of calcium-hydrogencarbonate type and not to the mineral sulphate water type.

Furthermore, surface flows in the area of central Dalmatia, i.e. the water from the watershed of the Cetina, Jadro and Žrnovnica Rivers is typical hydrogencarbonate water and does not belong to sulphate water.An exception is the small Slana Jaruga spring in Glavica near Sinj. Karst water, unlike mineral water which contains significant quantities of sulphates, can be found in the watershed of the Krka and Čikola Rivers and in the Imotski, Vrgorac and Baćina Lakes regions. It is not likely that they can reach the Split spa spring.

Seawater which is a constituent part of the sulphur water has a high sulphate content for the reduction and creation of hydrogen sulphide. Temperature plays an important role in the formation of

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H_2S. According to results of investigations, H_2S can be created if the seawater temperature ranges from 17-19°C.

The average temperature of the spring water with a normal content is 21°C. It does not oscillate greatly; the highest temperature it reaches is ca 24°C (Table 3). During stormy rainfall, the inflow of surface water into the spring increases and the temperature of that inflow is lower. In this phase oxygen appears and hydrogen sulphide disappears. This is known as the oxidation phase and it can last from several days to several months. According to Buljan, the normal sulphur phase has the following characteristic parameters: at least 10-14‰ chlorides, temperature ranging from 17-19° C, the presence of sulphur and oxygen.

The average temperature of the Split spa water with a normal content is ca 21°C; it does not oscillate greatly; the highest temperature it reaches is ca 24°C (Table 3). Balneotherapeutic action of the mineral water from the Split spa is based upon the influence of hydrogen sulphide. The therapy with mineral water in the Split spa is effected together with other procedures and methods of physical therapy and rehabilitation. This water is most often recommended for treating rheumatic illnesses. This water also alleviates pain in patients with psoriasis, sclerodermia and rheumatoid arthritis (Rosa, 1995; Krammer, 1961; Hercogova, 2002). It is also recommended for workers exposed to the influence of heavy metals such as printers, explosive experts and particularly for miners extracting mercury, since hydrogen sulphide binds heavy metals and has a detoxicating effect.

6. Conclusion

- The sulphur water from the Split thermal spa is, according to its chemical content, very similar to seawater but it has a lower degree of mineralization and contains hydrogen sulphide.
- Under the influence of great quantities of rainfall during cold periods (winter and beginning of spring) the ratio between seawater and surface water changes and thus the chlorides content and the content of other minerals are reduced and the lowered temperature reduces hydrogen sulphide which can disappear completely. A concentration of 12‰ chlorides (76-94.4% days / year) and 12 mg/L hydrogen sulphide (66.7 - 88.9% days/year) has been taken as a limit value between water with a normal typical content and water in the cold rainy periods.
- Occasionally, after long dry periods in winter and after the lowest sea level, the spring dries up for a short period. Then the discharge is decreased to 1/5 of the average spring capacity and the water inflow to the spring consists of hydrogencarbonate type surface water.
- According to the results of investigations carried out at the Split spa, the spring water in the dry period consists of surface, karst hydrogencarbonate water; this differs from past assumptions according to which sulphur water normally consists of seawater and mineral sulphate water.
- Since the hydrogen sulphide concentration in the Split water varies, it is necessary to monitor it constantly since different concentrations of hydrogen sulphide influence the treatment of dermatological diseases and joints.

Table 1. Results of the chloride and hydrogen sulphide examination in the spring of Split spa

Parameter	1952/53 N=49	1987 N=72	1988 N=72	2003 N=72
Chloride				
% of days above 12‰	85.7	76	80.6	94.4
$\overline{C}$	15.82	16.52	17.74	17.73
σ	1.31	1.41	1.81	1.26
CV, %	8.32	8.53	10.2	7.1
Range	1.33-19.5	2.9-17.92	0-19.95	1.27-19.65

% of days below 12‰	14.3	24	19.4	5.6
Month below 12‰	12,1	3,4,5	1,2,3	6
Hydrogen sulphide % of days above 12 mg/L $\bar{C}$ σ CV, % Range	 73.46 21.1 4.81 22.8 0-33.26	 73.6 20.11 3.07 15.26 0-23.7	 66.7 20.4 3.76 18.43 0-33	 88.9 18.9 2.76 14.60 7.4-23.9
% of days below 12 mg/L	26.53	22.4	33.3	11.1
Month below 12 mg/L	4,12,1,2	3,4,5	1,2,3,4	6
Rain-average mm from 1-3 months from 4-6 months from 7-9 months from 10-12 months		 113.43 56.5 34.56 55.17	 86.93 64.26 29.3 66.3	 93.73 50.7 34.13 73.83

N= number of analyses
$\bar{C}$=mean concentration
σ=standard deviation
CV=variation coefficient

Table 2. Composition of water from the spring intake structure and from the water flowing channel (2003)

Parameters	Water from the spring intake structure			Water from the water flowing channel		
	mg/L	mmol/L	eq/L	mg/l	mmol/L	eq/L
Temperature (°C)	16			19		
Hydrogencarbonate (HCO_3^-)	125.7	2.06	2.06	128	2.09	2.09
Chloride (Cl^-)	92	2.59	2.59	4437	124.98	124.98
Sulphate (SO_4^{2-})	32	0.333	0.666	715	7.447	14.895
Calcium (Ca^{2+})	83.2	2.08	4.16	188	4.7	9.4
Magnesium (Mg^{2+})	3.84	0.16	0.32	231	9.63	19.25
Hydrogen sulphide (H_2S)	0			0		

Table 3. Temperature in the spring of Split spa (1987, 1988 and 2003)

Month/year/date	1987	Temperature (°C)	1988	Temperature (°C)	2003	Temperature (°C)
January	12	14.3	10	14.4	20	14.0
February	2	16.4	5	14.9	5	15.9
March	10	19.9	15	19.5	11	18.9
April	5	20.6	3	20.2	8	20.0
May	4	21.1	2	21.0	10	21.5
June	2	20.58	5	20.0	5	21
July	22	21.5	20	20.9	15	21.5
August	5	21.6	4	21.4	8	21.5
September	15	22.7	11	23.5	20	23
October	30	23.75	20	22.9	23	23.2
November	12	20.9	10	20.5	16	20.5
December	8	21.1	10	21.5	18	20.2
Mean value ($\bar{C}$)		20.34		20.05		19.96
Standard deviation (σ)		2.58		2.77		2.58
Variation coefficient		12.7		13.8		12.9

(CV, %)						
Range		14.3-23.75		14.4-23.5		14.0-23.2

Table 4. The content of seawater and sulphuric water in percentages

Parameter	Sea water according to Buljan (1962)		Sulphuric water from the Split spa according to Miholić (1952)
Cation, %	Na	30.4	30.75
	K	1.19	1.28
	Ca	1.16	1.97
	Mg	3.7	3.5
Anion, %	Cl	55.2	54.46
	SO_4	7.7	7.76
	HCO_3	0.35	0.25

Table 5. The content of sulpuric waters in Croatia

		Split spa	Istrian spa	Varaždin spa	Tuhelj spa
Parameters g/kg					
Cations	Na	8.79	0.5889	0.1031	0.0109
	K	0.3661	0.0235	0.0096	0.0029
	Ca	1.00	0.4120	0.1286	0.0637
	Mg	0.5631	0.09422	0.0248	0.038
Anions	Cl	15.58	1.332	0.081	0.0026
	SO_4	2.20	0.0037	0.1499	0.00394
	HCO_3	0.144	0.5455	0.4728	0.371
Undissociated	SiO_2	2.94	0.0360	0.04682	0.0548
	Al_2O_3	3.08	0.0005	0.0008	
	Fe_2O_3	1.06	0.0014	0.003	
Mineralization H_2S, mg/L		28.67 20.0	3.427 23.3	1.1724 7.0	0.5841 7.6
Water classification		Hypothermal 21,3°C	Hypothermal 30,5°C	Hyperthermal 57,6°C	Hyperthermal 33,1°C
		Sulphuric	Sulphuric	Sulphuric	Sulphuric
			Radioactive		
Dominant cations>20 meq %		Na	Na, Ca	Ca, Na	Ca, Mg
Dominant anions>20 meq %		Cl	Cl	HCO_3, SO_4	HCO_3

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