Mineral and Thermal Waters in the Croatian Part of the Pannonian Basin

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Abstract Favourable geothermal properties which are characteristic of the major part of the Pannonian Basin System also extend into its south-western margin where Croatia is situated. Owing to a thin lithosphere, the geothermal heat flow is high, which enables groundwater to heat up. Some of the natural geothermal springs have a millennial tradition in different modes of utilization. The mode of utilization varies according to the temperatures, e.g. waters of lowest temperatures (17–20 °C) are used for fish farming, while waters of the highest temperatures (68–98 °C) are utilized for space heating and hot water preparation. In total, thermal waters in Croatia are utilized in the following ten activities: recreation, balneotherapy, water heating, space heating, greenhouse heating, fish farming and directly as sanitary water, public water supply and bottled table and mineral water. According to the major ionic composition, waters belong to the NaCaMg–HCO_3SO_4, CaMgNa–HCO_3SO_4, CaMgNa–HCO_3, CaMg–HCO_3, NaCa–HCO_3 or Na–Cl-type. Despite significant potential confirmed through multiple professional and scientific studies, the utilization of both thermal and mineral waters remains at low levels and traditional applications.

Keywords Mineral waters · Thermal waters · Geothermal properties · Hydrogeochemical properties · Utilization · Pannonian basin · Croatia

Introduction

The existence of thermal water springs in Croatia can be traced via the incidence of toponyms *toplica/e*, meaning hot water spring/s, and *topličica*, meaning warm water spring (diminutive refers to lower water temperature). Some of the localities have been utilized even in prehistoric time (Šimunić 2008). Before the Roman Empire, a

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number of Illyrian tribes populated areas of present day Croatia. One of the tribes occupied specifically the north-western part, with 25 natural warm and hot springs. They became known as *Iassi* all around ancient Europe (Schejbal 2003). Their name derives from the Greek root –ias/–iatria, meaning cure, because they were medicine–men using hot water healing powers. The same root is present today in the form iatros—physician (e.g. paediatrician). This fact shows that even before the arrival of the Romans, thermal springs had a specific place in the cultural landscape of the area. During the Roman Empire they were curative destinations, especially *Aquae Iassae* (Varaždinske toplice), *Aquae Balissae* (Daruvarske toplice), *Aquae Vivae* (Krapinske toplice), *Aquae Vitae* (unknown location) and *Ad fines* (Topusko), with many archaeological remains standing to this day. Mineral springs can also be recognized through toponyms. Highly mineralized cold springs are deemed Slatina (literally salty spring) and Kiselica (sour spring), referring to high dissolved CO_2 gas. They have been utilized for drinking and bottling for centuries as well.

Since thermal and mineral waters have been utilized during a vast time period in the present-day Croatian territory, it is clear that researchers were devoted to their exploration. The oldest chemical analyses date back to the last decades of the 18th century (Crantz 1777). In that period, mostly chemists and medical doctors were showing interest in the subject due to applications in balneology. Comprehensive geological research started at the end of the 19th century (Pilar 1884; Koch 1889; Voyt 1890), but reached its peak during the 1970s oil crisis. At that time the Federative Republic of Croatia (then a part of Yugoslavia) established a fund dedicated specifically to the exploration of thermal and mineral water. Major research results were summarized in a monograph "Geothermal and mineral waters of the Republic of Croatia" (Šimunić 2008), to avoid unnecessary repetition of work, since it provides a broad overview of the existing bibliography. The possibilities of geothermal energy utilization were analysed in the Geoen-Programme of geothermal energy utilization (EIHP 1998), as well as in a number of professional and scientific publications mentioned later in the text.

The aim of this paper is to give an overview of the occurrences of thermal and mineral waters in the Pannonian part of Croatia, their characteristics, utilization, sustainability issues and predictions of future development.

Geological Settings

Croatia is situated at the junction of major European regions: Alps, Dinarides and the Pannonian Basin System (PBS) (Fig. 1a). It is divided distinctively into two parts: the Pannonian part in the north–east and the Dinaridic part in the south–west (Fig. 1b). The north-eastern part of Croatia represents the south-western margin of the Pannonian Basin System (PBS), and the majority of Croatia's geothermal potential is concentrated there. It is characterized by high average geothermal gradient (49 °C/km) and surface heat flow (76 mW/m^2) (EIHP 1998). Conversely,

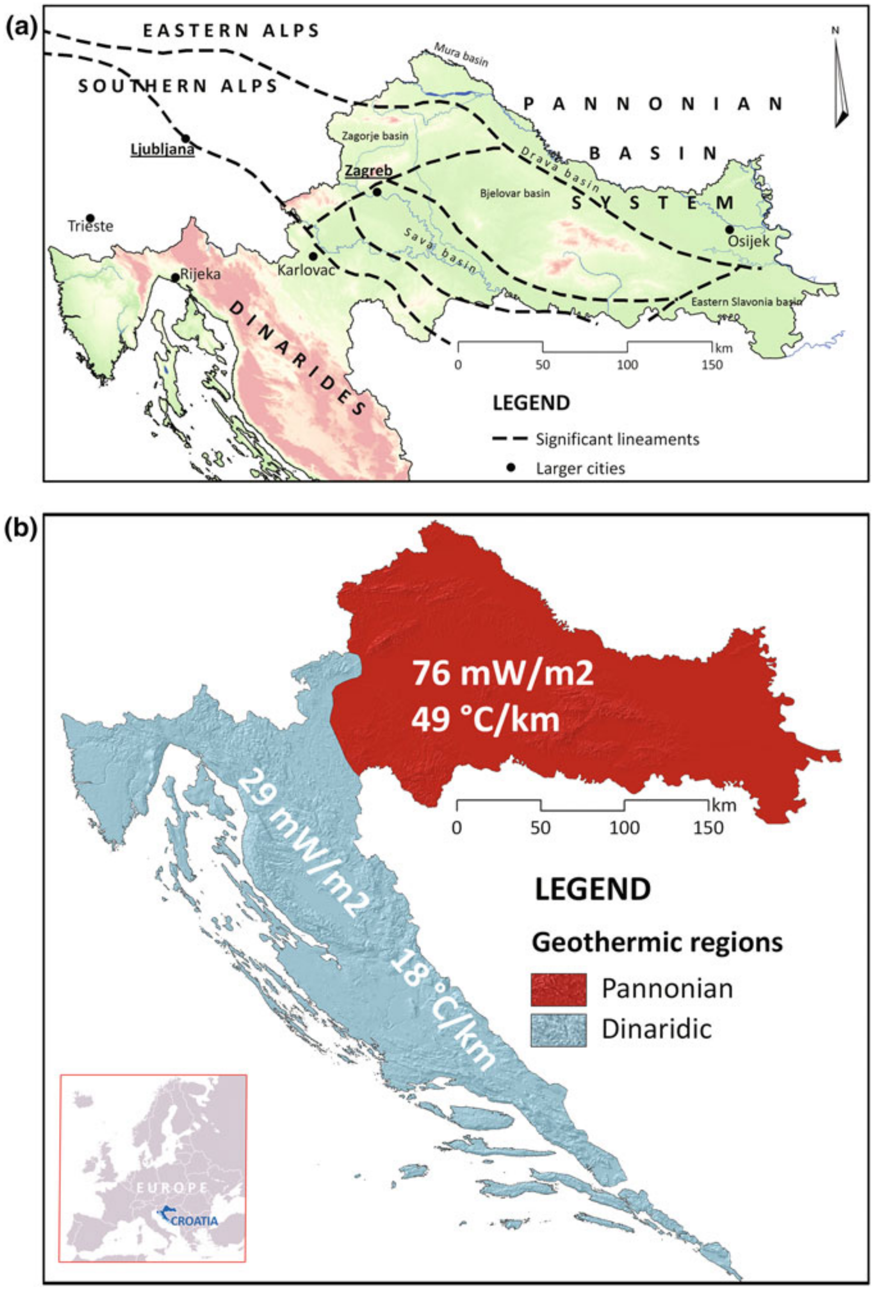

Fig. 1 **a** Position of Croatia in relation to major European tectonic units (according to Tari and Pamić 1998; Lučić et al. 2001; Velić et al. 2012), **b** heat flow density and geothermal gradient in geothermically different Croatian regions

the Dinaric part has low average geothermal gradient (18 °C/km) and surface heat flow (29 mW/m^2) (EIHP 1998).

Differences in geothermal traits are caused by the regional tectonic setting. The Mohorovičić seismic discontinuity (Moho), a boundary between the Earth's crust and mantle, is deepin the Dinarides—50 km, and only 28 km in the Pannonian part (Aljinović and Blašković 1984). Since the mantle convection transports heat more efficiently than conduction in the crust, places where the mantle is closer to the surface will experience higher geothermal heat flow. High values of geothermal parameters in the Pannonian Basin System are a consequence of the middle Miocene (≈16–11.6 Ma b.p.) back-arc extension of the basin, which led to lithosphere thinning and enabled hot asthenosphere to approach the surface (Horváth et al. 2015).

Attempts to explain the occurrence and origin of thermal springs in Croatia date back as far as the 18th century. Many Croatian geologists of those times were studying the warm springs and postulated a number of different hypotheses. Gorjanović-Kramberger (1904) was convinced that thermal springs in north-western Croatia were of volcanic origin, with magma chambers heating the waters and deep faults, *thermal lines*, bringing them to the surface. The main shortcoming of this hypothesis is that there are no recent magmatic bodies to supply heat, but because of Gorjanović-Kramberger's authority it has supporters even today (Šimunić 2008). The idea that thermal waters have meteoric origin, and then get heated during circulation through the underground, first appeared in the works of Pilar (1884). Afterwards, geochemical research of Miholić (1940, 1952, 1959), Horvatinčić et al. (1991, 1996), Marović et al. (1996), Marković and Kovačić (2006), Bituh et al. (2009), Polančec (2011), Kapelj et al. (2014) and Marković et al. (2015) proved that thermal waters of the area are of meteoric origin. As the understanding of the structural fabric of Croatian territory had been advancing, it was proven that the majority of natural thermal springs in the Pannonian part of Croatia are situated in the intersections of anticline crest sand transverse faults (Šimunić and Hećimović 1999). This kind of environment is usually highly fractured and a stark permeability contrast to the surroundings enables the upwelling of heated water from depth to the surface (Caine et al. 1996; Curewitz and Karson 1997; Evans et al. 1997; Faulkner et al. 2010). Water temperature is dependent on the fault dip: if it is vertical, the water loses less heat on the way to the surface, which results in warmer springs. The more the fault deviates from vertical, the more heat is lost on its way to the surface, resulting in ever lower spring temperatures. When the conduits are sub-horizontal, the water can cool down but retains high mineralization and in such cases mineral springs occur (Šimunić 2008).

Mineral and Thermal Water Locations and Categorization

Croatian thermal localities are subdivided into two categories: springs and deep boreholes. Springs are localities where thermal water naturally flowed out or is still flowing out from the aquifer onto the surface. In time, if larger quantities were needed,

Table 1 Categorization of geothermal localities in Croatia on the basis of water temperature (Borović and Marković 2015)

Category	Subthermal	Hypothermal	Homeothermal	Hyperthermal
T (°C)	13–20	>20–34	>34–38	>38
Natural spring	1	6	2	6
Deep borehole	0	4	0	7

intake structures or shallow boreholes were made. At some of those localities natural springs have dried up following higher pumping rates. The category of deep boreholes accounts for all the localities where there are not, nor have there ever been natural springs, and thermal waters were found during hydrocarbon exploration.

Except by the mechanism that they come to the surface, thermal waters also differ in their temperatures. In Croatia they are categorized according to the modified balneological classification (Table 1), created on the basis of traditional Croatian balneological classification which existed since the 1950s (Haramustek et al. 1952), but actually stemmed from considerations of German and Swiss medical balneology experts at the beginning of the 20th century (Jacobj 1907; Hintz and Grünhut 1916; Hartmann 1925).

It is visible that the point of reference for this scale is the average human body temperature, so homeothermal means the same as human temperature (from Greek *homós*—the same), hypothermal is below that temperature (Gr. *hypó*—below) and hyperthermal is above body temperature (Gr. *hypér*—above). The modification was done in the lower part of the scale because in balneology waters with temperatures lower than 20 °C are not considered (Kovačić and Perica 1998). From the hydrogeological point of view, however, all groundwaters with temperatures higher than the average annual temperature of the locality are considered thermal, albeit they cannot be used in balneology. The temperature range in which waters are currently being utilized is from 17 to 98 °C. There are also some deep boreholes which yield waters of higher temperatures.

In Croatia, groundwater is usually characterized as mineral if the total dissolved solids (mineralization) is higher than 1 g/L, or the temperature higher than 20 °C, or contains higher concentrations of a specific dissolved compound that has a strong physiological effect, for example: Fe > 10 mg/L; F > 2 mg/L; J > 1 mg/L; S > 1 mg/L; As > 0.7 mg/L; CO_2 > 1 g/L; Rn > 81.4 Bq/L; Ra > 3.7 Bq/L (Tušar 1998).

Hydrogeochemical Characteristics

A compilation of geochemical data from different sources has been used in this paper to give a general overview of hydrogeochemical characteristics of thermal and mineral waters. It includes chemical analyses by Miholić (1952), Jurišić-Mitrović (2001), Brkić et al. (2007), water quality monitoring of hospitals for medical rehabilitation Varaždinske, Stubičke, Krapinske and Bizovačke Toplice,

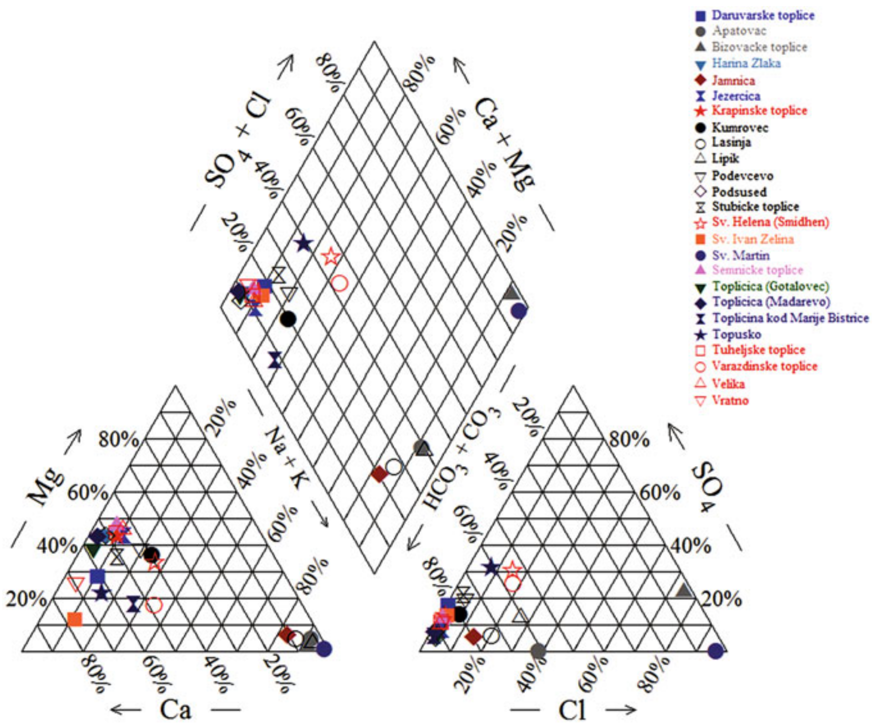

Fig. 2 Piper diagram of selected waters

and chemical analyses performed during research for the Basic Hydrogeological Map, Krapina and Varaždin sheets, in October 2009.

According to the major ionic composition, waters from Varaždinske toplice and Sv. Helena (Šmidhen) belong to a NaCaMg–HCO_3SO_4 mixed type; Podevčevo, Topusko and Stubičke toplice waters belong to a CaMgNa–HCO_3SO_4-mixed-type; Topličina kod Marije Bistrice belongs to a CaMgNa–HCO_3-mixed-type; Harina Zlaka, Tuheljske toplice, Krapinske toplice, Topličica (Mađarevo), Topličica (Gotalovec), Sutinske toplice, Šemničke toplice, Velika, Vratno, Sv. Ivan Zelina, Podsused, Jezerčica, and Daruvarske toplice, belong to the CaMg–HCO_3-mixed-type; Jamnica, Lasinja, Lipik and Apatovac belong to a NaCa–HCO_3-mixed-type and Bizovačke toplice and Sv. Martin waters belong to a Na–Cl-type (Fig. 2). Hydrochemical facies, as recognised from water chemistry data, are a consequence of the chemistry of the recharging water, and water-aquifer matrix interactions, as well as groundwater residence time within the aquifer.

The total dissolved solids (mineralization) are higher than 1000 mg/L (or 1 g/L) in the waters of Lipik, Jamnica, Apatovac, Lasinja, Sv. Martin and Bizovačke toplice (Fig. 3; Table 2). According to the mentioned mineral water classification, they are classified as mineral. Lower mineralization is observed in all other waters and it ranges from 372 mg/L; (Kumrovec) to 963 mg/L (Varaždinske toplice).

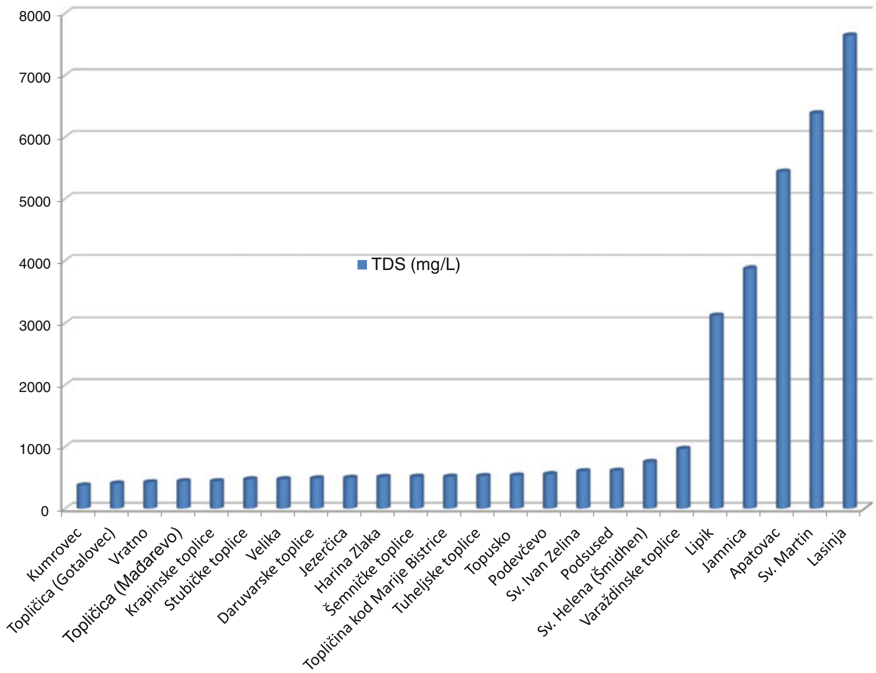

Fig. 3 Distribution of total dissolved solids

From the viewpoint of water temperature, all waters, except the water from Topličina kod Marije Bistrice, are mineral waters because they have temperatures higher than 20 °C (Fig. 4).

Furthermore, water quality monitoring of thermal waters of Varaždinske toplice, Stubičke toplice, Krapinske toplice, Bizovačke toplice and Lipik points to a high content of CO_2, or H_2S, or I^-. In the thermal waters of Varaždinske toplice, a large amount of H_2S (10.4 mg/L) has been observed (Special Hospital 2014). In the waters from Stubičke toplice and Lipik, a high content of CO_2 is registered—101.49 mg/L (Hospital Stubičke Toplice 2014) and 3500 mg/L (Šimunić 2008), respectively. In the waters from Bizovačke toplice and Krapinske toplice, H_2S is also measured (Bizovačke toplice 2014; Hospital Krapinske Toplice 2014). Waters from Bizovačke toplice and Sv. Martin have concentrations of iodide exceeding 1 g/L. Other thermal waters do not exhibit elevated concentrations of specific dissolved compounds.

Waters from Jamnica, Lasinja and Apatovac have a high CO_2 content, with concentrations ranging from 2289 to 3890 mg/L (Šimunić 2008). These waters also have high concentrations of sodium, potassium and chloride; concentrations ranged from 903.9 to 1905 mg/L (Na^+); from 14.6 to 106.8 mg/L (K^+); and from 394.6 to 1011.2 mg/L (Cl^-) (Table 2). The highest concentrations of sodium, potassium and chloride are present in waters from the localities Bizovačke toplice and Sv. Martin (Table 2).

Table 2 Mean values of observed geochemical parameters at localities

	T (°C)	HCO_3^- (mg/L)	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)	Na^+ (mg/L)	K^+ (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	TDS (mg/L)
Apatovac	12.1	2726.4	54.4	42.4	1583.9	14.6	1011.2	3.1	5436
Bizovačke toplice	90.0	949.3	503.6	79.5	8528.0	226.3	14,750	5926	30,963
Daruvarske toplice	48.2	314.3	77.7	21.7	13.2	3.8	2.0	52.9	486
Harina Zlaka	24.2	362.0	68.7	36.0	7.2	2.3	7.7	25.7	510
Jamnica	12.0	2334.9	106.8	37.7	903.9	106.8	256.8	127.8	3875
Jezerčica	38.4	346.1	58.0	34.0	8.5	15.1	7.8	24.9	494
Krapinske toplice	41.2	298.2	55.3	30.9	10.9	3.1	3.1	38.0	440
Kumrovec	25.0	234.9	40.1	22.4	23.9	7.9	10.6	31.9	372
Lasinja	12.5	4426.0	168.8	57.3	1905.0	76.3	724.1	281	7639
Lipik	52.6	1519.1	30.9	14.7	817.1	91.5	394.6	251.9	3120
Podevčevo	17.7	341.3	58.8	32.8	26.1	9.4	14.3	71.0	554
Podsused	30.5	446.2	82.2	42.5	9.6	1.4	7.8	20.0	610
Stubičke toplice	44.9	284.5	64.8	27.2	16.7	4.3	8.4	64.1	470
Sv. Helena (Šmidhen)	27.3	347.7	83.3	42.5	59.8	7.8	57.0	153.5	752
Sv. Ivan Zelina	21.4	394.5	118.0	11.2	19.3	2.0	7.0	49.7	602
Sv. Martin	36.8	137.0	45.2	15.9	3864.0	98.2	2217	5.0	6382
Šemničke toplice	32.7	355.6	61.5	39.0	9.7	3.0	3.6	40.6	513
Topličica (Gotalovec)	25.6	291.7	60.2	24.8	3.9	2.1	5.2	15.1	403
Topličica (Mađarevo)	23.0	320.1	61.2	29.9	3.3	1.4	3.3	20.5	440
Topličina–Marije Bistrice	17.8	358.2	72.5	14.2	36.3	9.6	4.3	17.9	513
Topusko	47.5	264.6	90.0	19.2	17.9	12.3	19.8	108	532
Tuheljske toplice	32.8	367.9	63.8	36.9	11.3	3.0	3.5	37.6	524
Varaždinske toplice	55.3	450.2	126.1	27.8	82.4	34.6	82.6	159.5	963
Velika	28.2	336.1	53.2	33.5	13.0	1.6	4.2	30.8	472
Vratno	22.0	280.5	83.1	18.8	5.3	2.2	4.0	29.2	423

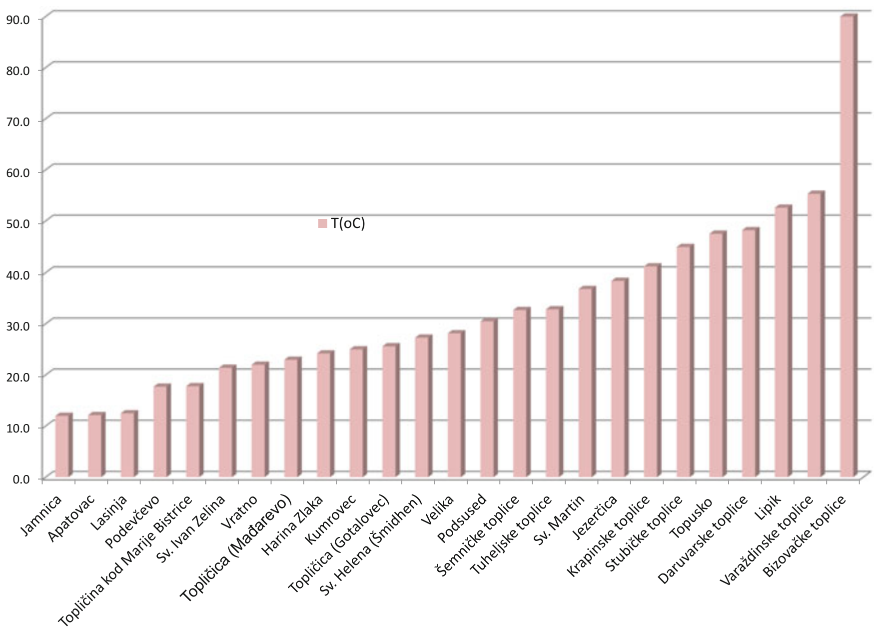

Fig. 4 Distribution of water temperatures

Utilization

The utilized waters considered in this overview range from subthermal to hyperthermal (Fig. 5).

The mode of utilization varies according to temperature, e.g. waters of the lowest temperatures (17–20 °C) are used for fish farming, while waters of the highest temperatures (68–98 °C) are utilized for space heating and hot water preparation. In total, thermal waters in Croatia are utilized in the following ten activities: recreation, balneotherapy, water heating, space heating, greenhouse heating, fish farming and directly as sanitary water, public water supply, and bottled table and mineral water (Table 3).

The most frequent modes of utilization are recreation and balneotherapy, which is the modus known from prehistoric times and antiquity. It is followed by water and space heating and utilization of thermal water for sanitary purposes.

Mineral/thermal waters from the localities Apatovac, Jamnica, Lipik and Topličica (Gotalovec) are utilized for water bottling, and from the localities Harina Zlaka and Vratno for public water supply.

Thermal water utilization in Croatia is stagnating, rather than experiencing growth. Variations in the number of users are mostly the result of temporary closures due to necessary renovation and retrofitting of the outdated infrastructure (Borović and Marković 2015). At some localities where geothermal water is

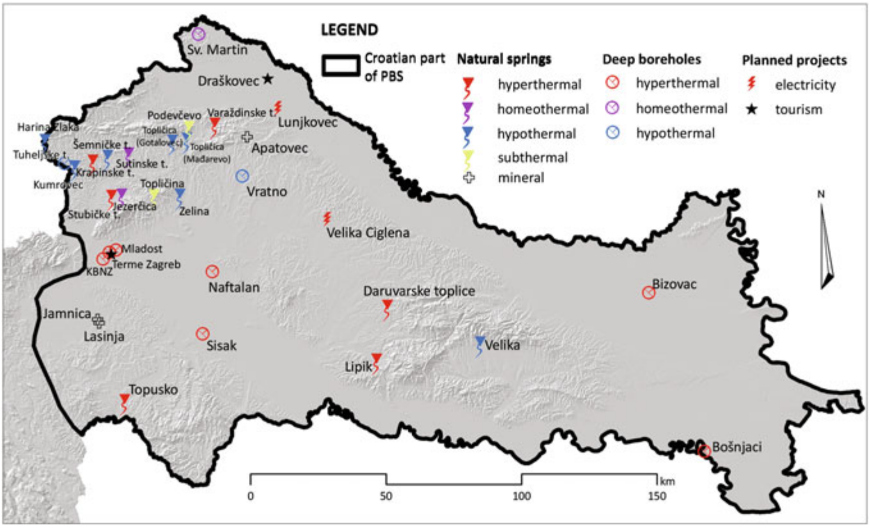

Fig. 5 Locations of thermal and mineral water utilization in the Croatian part of the Pannonian Basin System

Table 3 Geothermal water utilization in Croatia in the year 2014 (Borović and Marković 2015)

Utilization	Type of locality		
	Natural spring	Deep borehole	Total
Recreation	14	3	17
Balneotherapy	7	2	9
Water heating	2	3	5
Space heating	4	4	8
Sanitary water	5	1	6
Fish farming	2	1	3
Greenhouse heating	1	3	4
Table water	0	1	1
Mineral water	1	3	1
Public water-supply	1	1	2

currently being utilized in a single mode, there are plans to increase pumping rates and introduce new modes of utilization since there is a consensus that having more users and extracting as much heat as possible significantly increases economic viability of thermal projects (Rybach 2003; Legmann 2003; Lund et al. 2005; Pravica et al. 2006). The most diversified use is accounted for in Stubičke toplice (Fig. 5): recreation, balneotherapy, water and space heating, greenhouse heating and sanitary water.

Other than improving heat extraction at active locations, significant interest is present to utilize thermal waters and/or energy in new locations (Borović and Marković 2015). The projects in higher stages of development include Terme Zagreb (using a set of existing boreholes) and Draškovec (Međimurje County in the north of Croatia) (Fig. 5). For all the mentioned localities, the plan is to utilize thermal waters in cascade systems for facility heating, hot water preparation, spa and recreation, greenhouse heating and/or fish farming, to make the process economically feasible.

Borović and Marković (2015) clearly show that the potential of geothermal electricity generation in Croatia could not supply a significant portion of the country's electricity demands. It is considered marginally profitable, depending on the project, because available resources have temperatures up to 170 °C, i.e. low enthalpy sources, which are used for the less economical binary process (Kristmannsdóttir and Ármannsson 2003). The most significant potential for power generation was identified in Velika Ciglena and Lunjkovec (Fig. 5). According to the Croatian National Renewable Energy Action Plan (Ministry of Economy 2014), the first geothermal power plant Marija-1 (4.71 MW_e) is supposed to start operation in the year 2016 in Velika Ciglena. The projects also include cascade utilization of thermal water to make the schemes economically viable.

The Croatian heating and cooling sector could benefit greatly from large scale geothermal utilization, since full development of the existing fields could supply over 13 % of the energy demand, excluding geothermal heat pump utilization (Borović and Marković 2015). There are 3500 boreholes in the Pannonian part of Croatia, leftover from hydrocarbon exploration and exploitation, many of which represent an untapped local energy micro potential (Kolbah and Škrlec 2010). The idea of utilizing these boreholes is neither new, nor unexpected. It has been determined that significant energy potential is present in the water surrounding mature hydrocarbon extraction sites with deep boreholes, high temperatures and favourable permeability conditions, which could be retrofitted for geothermal water extraction during and after hydrocarbon exploitation (Čubrić 1978; Kurevija and Vulin 2011). One of the obstacles for stronger integration of geothermal energy into the heating sector is the fact that the same region rich in geothermal potential also has a developed gas pipeline network. Natural gas heating is a strong competitor because it is readily available, while geothermal boreholes are often a few kilometres away from settlements, so it would be necessary to build insulated hot water pipelines, which is very costly (Čubrić 1993; Fridleifsson 2003; Szita 2015). In this situation, even when awareness of possible renewable resource utilization exists, it succumbs to economic interests.

Sustainability and Environmental Impacts of Thermal Water Utilization

The need for protection was actually the very thing that prompted early research of geothermal springs in Croatian territory, after the Croatian parliament passed the law on "*Protection of curative and mineral baths*" in 1885. Protection zones around the springs were quite extensive and there are multiple court disputes over possibilities of private land management and digging of drinking water wells in protective zones conserved in documentation of that time (Šimunić 2008). Scientists and the authorities were obviously aware of the potential danger that nearby boreholes could present to natural springs. Geological research has since demonstrated that recharge of geothermal aquifers, which feed the springs, occurs in mountainous hinterlands of the springs (Šimunić and Hećimović 1999). This means that the danger to such springs comes also in the form of forestry activities and large quarries. The utilisation of explosives is particularly dangerous because it creates seismic events that can impair hydraulic properties of both the aquifer and the discharge area. The troubling fact is that the dominant geothermal aquifers in Croatia are Triassic dolomites which are also being mined in 17 active quarries in the Pannonian part of the country (Živković and Krasić 2008). There were many reports from Sutinske toplice (Fig. 5) that mining in the nearby dolomite quarry has the same impacts as natural earthquakes, i.e. changes in yield of the springs. More drastic was the example from nearby Šemničke toplice (Fig. 5), where the spring did actually dry up after intensive mining in the same quarry, only to reappear after a longer period of time, but in two new locations (Šimunić 2008).

Waters from deep confined aquifers discovered during hydrocarbon exploration are not a part of the contemporary hydrologic cycle, meaning they are devoid of any significant recharge and can only be utilized in a sustainable manner by production–reinjection doublets (Eckstein and Eckstein 2005). Aside from that, their quality can be reduced by oilfield waste disposal in the boreholes, which has become a frequent practice (Šimunić 2008).

Discussion and Concluding Remarks

Favourable geothermal properties, which are characteristic of the major part of the Pannonian Basin System, also extend into its south-western margin where Croatia is situated. Owing to a thin lithosphere, geothermal heat flow is high and enables groundwater to heat up. Some of the natural geothermal springs have a millennial tradition in different modes of utilization. In the 20th century new thermal and mineral water resources were identified during hydrocarbon prospecting. According to temperature, thermal waters are predominantly hypothermal and hyperthermal, both at natural springs and boreholes. The majority of thermal waters have low mineralization and a low CO_2/H_2S content and they can be utilized as potable water, which is already the case at some locations. Others have high mineralization and a high CO_2/H_2S content. Mineral waters have high dissolved CO_2 and a high content of sodium, potassium, and chloride. The type of thermal and mineral water occurrence and the modes of utilization dictate the potential problems that did or could arise. In the Croatian example, the vulnerability of utilized sources of geothermal water to qualitative and quantitative status deterioration differs among natural springs fed by hydrothermal systems and waters from deep boreholes, since the former are part of the contemporary hydrologic cycle, while the latter are not. In accordance with their characteristics, natural springs are mainly endangered by forestry and quarries, while waters from deep boreholes are at risk of contamination by fluid waste disposal. Despite significant potential confirmed through multiple professional and scientific studies, the utilization of both thermal and mineral waters remains at low levels and traditional applications.

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