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GEOME 2
Manuscript received: September 15, 2024
Accepted: November 6, 2024

Geologica Macedonica, Vol. **38**, No. 2, pp. 131–139 (2024)
On print ISSN 0352 – 1206
On line ISSN 1857 – 8586
UDC: 556.36:551.435.8]:550.4(497.762)
https://doi.org/10.46763/GEOL24382131g

Original scientific paper

NATURAL TRACERS AS A TOOL FOR UNDERSTANDING THE FUNCTIONING OF THE KARST SYSTEM AND ITS VULNERABILITY – A CASE STUDY OF THE MANASTIREC SPRING –

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A b s t r a c t: The Manastirec spring is the only drinking water source that supplies 200 inhabitants of the municipality Goren Manastirec in west-central Macedonia. Natural tracers were applied in order to characterize the properties of the ground water flow and transport of contaminants through the karst system and to study the functioning of the spring. Water level, temperature and electrical conductivity of the spring were measured with data logger in hourly intervals during several rain events in 2015 and 2016. Rain events caused rise of water level and decrease of both water temperature and electrical conductivity. Concentrations of calcium, magnesium, sulphate and nitrate present in spring water were analyzed during the end of July 2016, during three rain events. The results showed that sulphate and Ca/Mg ratio increase, whereas nitrate decrease after rain events. All results demonstrate high vulnerability of the Manastirec spring to contamination.

Key words: natural tracers; hydrochemistry; Manastirec karst spring

INTRODUCTION

Ground water of karst aquifers is very important source of drinking water supply. At the same time, karst aquifers are commonly highly vulnerable to contamination and are impacted by a wide range of human activities as urban settlements, industry, agriculture and traffic (Drew & Hötzl 1999; Kogovšek, 2011; Petrič et al, 2011). In order to protect the karst water from pollution it is necessary to investigate the properties of the ground water flow and transport of contaminants through the karst systems.

Different research methods are used in the studies of karst water. Artificial and natural tracers are often applied to study the hydrogeological functioning and response of karst aquifers. Physical properties of water, such as temperature and electrical conductivity, are tools that are often used as natural tracers. Simultaneous consideration of natural tracers and spring discharge (water level) provides information about the transfer functioning of the karst system (Petrič, 2002; Ford & Williams, 2007). This kind of analysis is often applied in the world (Gabrovšek & Peric, 2006; Nguyet & Goldscheider, 2006; Mulec et al., 2019), and in the Republic of Macedonia they are solely for the karst spring Slatinski Izvor (Gičevski et al., 2016).

An important characteristic of karst water is the prominent role of hydrometeorological events (flood events) on chemical changes in the aquifer. Therefore any hydrochemical information needs to be coupled with a hydraulic information (discharge, water level) (Perrin, 2003). Hydrochemical methods help to locate and quantify the mineralization of the water, provide information about the structure of the aquifer, the residence time of water and the various conditions of circulation that coexist within them (Hunkeler & Mudry, 2007; Lopez-Chicano et al., 2001).

The Manastirec spring is the only natural resource for drinking water of the Goren Manastirec municipality. This study is based on natural tracers that are an important tool for investigating the functioning of the karst system and assessment of its vulnerability to pollution.

Water level, temperature and electrical conductivity of the spring, as well as concentrations of calcium, magnesium, sulphate and nitrate present in water were measured during several rain events in 2015 and 2016.

STUDY AREA

The Manastirec spring (Figure 1) is located in the basin of Poreče in west-central Macedonia, in the valley of the Manastirečka Reka river.

The wider area around the source belongs to two geotectonic units: the Pelagonian Horst Anticlinorium and the West Macedonian Zone (Arsovski, 1997). It is built of Precambrian, Riphean-Cambrian, Paleozoic, Mesozoic, Tertiary, and Quaternary rocks (Figure 2) (Dumurdzanov et al., 1978, 1979). The Manastirec spring belongs to the Pelagonian Horst Anticlinorium (Figure 2).

The oldest rocks in the Pelagonian Horst Anticlinorium are Precambrian dolomite marbles that are tectonically crushed and well karstified, and graphite and quartz-muscovite schists of Riphean-Cambrian age.

Fig. 1. The karst spring of Manastirec (photo: B. Gičevski)

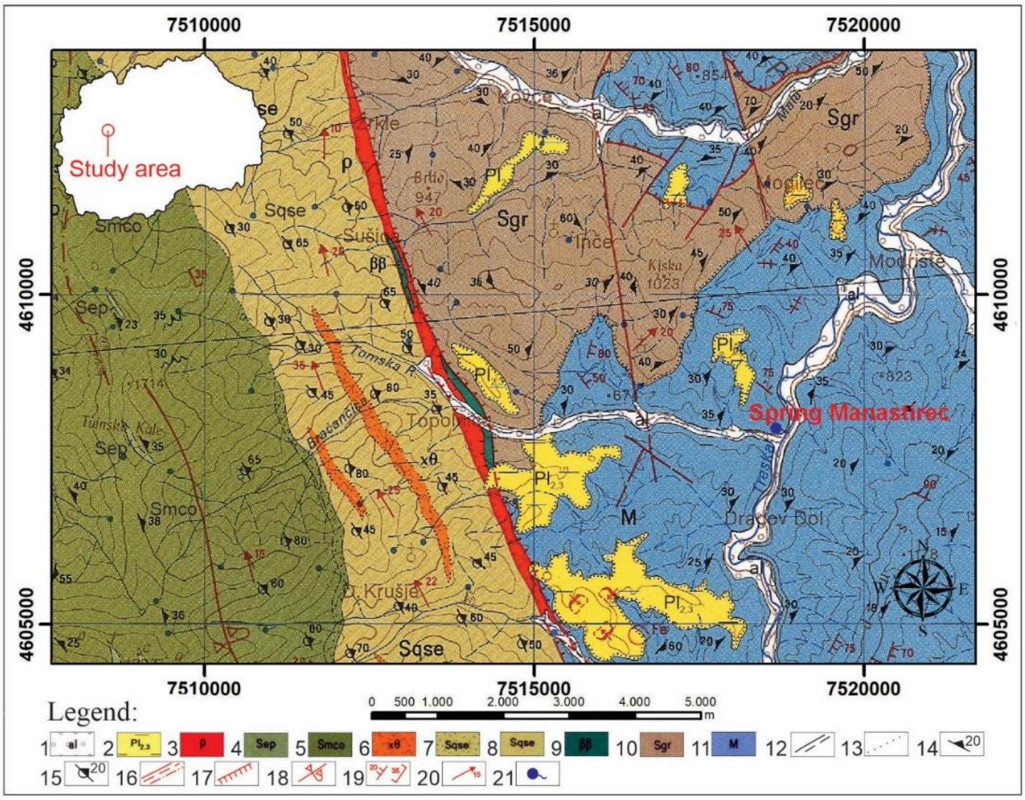

Fig. 2. Geological map of the researched area (Dumurdzanov et al. 1978)
Legend: **Quarter:** 1. **A**lluvium; **Tercier:** 2. Pliocene sediments (gravel, sands, clay); **Mesozoic:** 3. Aplitic granite; **Paleozoic**: 4. Epidote-chlorite-amphibole schists, 5. Muscovite-chlorite-quartz schists, 6. Metariolite, metariolite tuffs, 7. Feldspatic quarz-sericite schists and metasanstones, 8. Quartz-sericite schists and metasandstones, 9. Metadiabase; **Precambrian:** 10. Graphitic and quartz-muscovite schists (Riphean-Cambrian), 11. Dolomite marbles, 12. Normal border established and assumed 13. Gradual transition established, 14. Foliation normal, 15. Foliation inverse, 16. Faults, 17. Active contact, 18. Anticline axis leaning, 19. Crack and vertical crack, 20. Elements of axis drop of small folds, 21. Spring

The West Macedonian zone is built of Paleozoic epidote-chlorite-amphibole schists, muscovite-chlorite-quartz schists, metriolilites, metariolite tuffs, quartz-sericite schists and metasandstones. These are moderately permeable rocks with fissured porosity.

The contact between the two tectonic units is composed of Paleozoic metadiabases and Mesozoic aplite granites that have fissure porosity. The dolomite marbles are covered with Pliocene sediments (gravel, sand, clay) which in some parts are well-permeable rocks and they have intergranular porosity. Quaternary sediments are represented by alluvium, which have intergranular porosity, and fill the riverbed of the Manastirečka Reka river.

The two tectonic units are separated by a fault oriented in the NW-SE direction that is tracked for more than 50 km (Dumurdzanov et al., 2005). There is a fault between the villages of Slatina and Zrkle in the NW-SE direction, and several faults in the NW-SE direction in the lower parts of the Manastirečka Reka river valley.

The sources of the Manastirečka river and its tributaries Tomska Reka and Bračančica are in a non-carbonate environment. In contact with carbonate rocks, the Manastirečka river sinks underground as a point input into the karst system and flows underground. As a result, the riverbed is dry in summer.

Hydrogeological characteristics of the wider environment of the Manastirec spring are shown on the hydrogeological map (Figure 3). According to the structural type of porosity which occurs in rocks that are present within the wider environment of the investigated field can be distinguished: boundary type of aquifer, karst aquifer, and fissure type of aquifer.

Boundary type of aquifer is formed in Quarter alluvial sediments and Tercier Pliocene sediments (gravel, sands, clay), in which it is present intergranular porosity. Alluvial sediments are developed in the river of Manastirečka Reka.

Fissure type of aquifers is formed in Mesozoic aplitic granite, Paleozoic epidote-chlorite-amphibole schists, muscovite-chlorite-quartz schists, metariolite, metariolite tuffs, feldspatic quarz-sericite schists and metasanstone, quartz-sericite schists and metasandstones, metadiabase, and Riphean-Cambrian graphitic and quartz-muscovite schists.

Karst aquifer has developed in karstified Precambrian dolomite marbles, characterized by fracture – cavernous porosity.

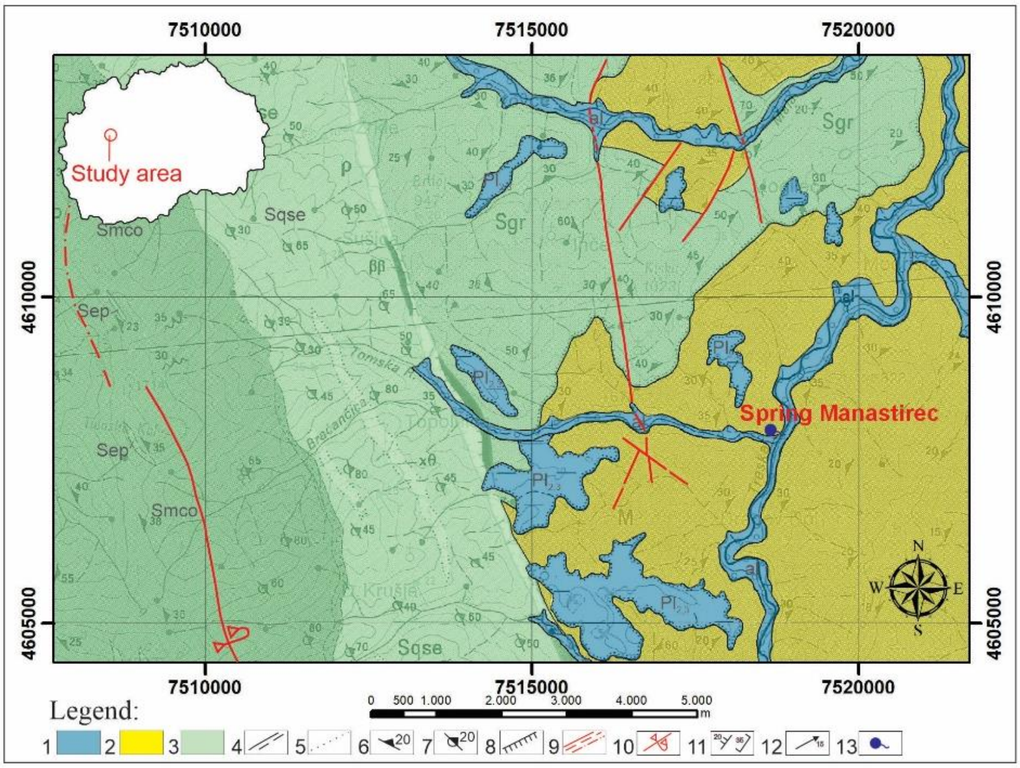

Fig. 3. Hydrogeological map of the researched area, according to Mirčovski.
Legend: 1. Boundary type of aquifer; 2. Karst aquifer; 3. Fissure type of aquifer

METHODS AND MATERIALS

Electrical conductivity (EC), water temperature and water level of the Manastirec spring were carried out in the period 2015–2016 using an Eijkelkamp CTDI 271 data logger, in hourly intervals. The relation between water level data and series of parallel measurements of levels and discharges was used for the estimation of discharges of the Manastirec spring.

We used data from a rain gauge in the village Lokvice which measured precipitation at daily intervals.

The chemical properties of water were analyzed at the Institute of Biology, Faculty of Natural Sciences and Mathematics, Ss. Cyril and Methodius University in Skopje. All water samples were filtered within 12 hours of collection and analyzed within 3–4 days. Sulphate was determined by photometric method (Dévai et al., 1973). The major cations calcium (Ca^{2+}) and magnesium (Mg^{2+}) were analyzed by wet digestion followed by atomic absorption spectrometry on Agilent 55Z or graphite furnace Agilent 240Z (Allen, 1989). All of the values are presented in mass concentrations (mg·l^{-1}). The mole ratio of calcium and magnesium ($r = n(\mathrm{Ca}^{2+})/n(\mathrm{Mg}^{2+})$) was calculated based on the values of the respective ions expressed in meq·l^{-1}.

RESULTS AND DISCUSSION

Rapid changes in discharges and changes in the physical properties and chemical composition of the water are characteristic of karst springs. Because of this, to study the functioning of the karst system numerous field measurements and data are necessary, more than for non-karst terrain.

Hydrograph of karst springs are indicators of karst aquifers (Bonacci, 1993) and they represent the result of various processes taking place in the karst basin from the moment when the precipitation fell on the basin to the moment when it reached the spring outlet (Kresic and Bonacci, 2010). Discharge hydrograph mostly of descending springs draining heavily karstified aquifers with numerous large karst conduits react very quickly to the precipitation that falls on the basin, whereas, deep, mostly rising karst springs, isolated from the influence of the processes that take place in the catchment areas, react more slowly (Bonacci and Roje-Bonacci, 2023). Any variation in electrical conductivity and water temperature in various time increments indicates that the water comes from another part of the aquifer or basin, that it is transported quickly through large karst conduits or slowly through the karst matrix, and that it stays in the karst aquifer for a shorter or longer time. Seasonal and daily variations of karst spring water's geochemical and physical parameters characterize the karst aquifer's different types and functioning (Benderitter and Roy, 1993; Birk et al., 2004; Pratama et al., 2021).

Flood pulses from rain events in 2015 and 2016

Time interval between September 9th and September 12th, 2015

This time interval was selected at the end of a longer dry period. In the period between August 18th and September 9th, 2015, in the area, only 1.5 mm of rain had fallen. During this period, the water level varied from 44.26 to 49.47 cm, the water temperature was between 11.9 and 12.09 °C, and EC oscillated between 480 and 492 μS/cm.

The intensive rains occurred on September 10th and 11th (29 mm and 10 mm, respectively) which caused an increase of water level (Figure 4).

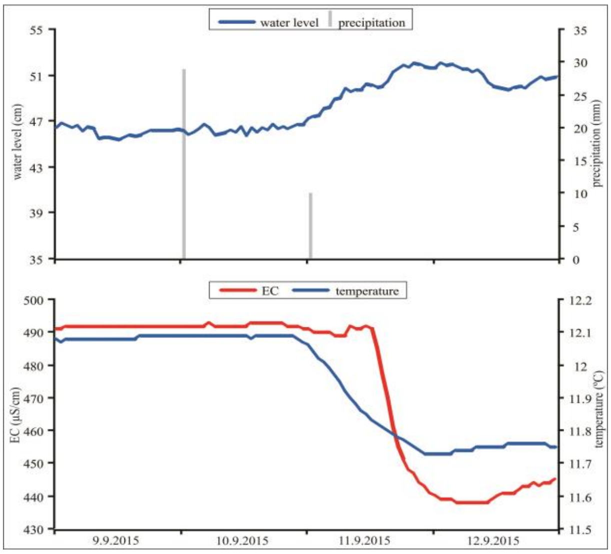

Fig. 4. Measurements of precipitation, water level, temperature and electrical conductivity (EC) in the period between September 9th and September 12th, 2015, every hour

Maximal level was reached on September 12th at 1:00 (52.02 cm). During the water level increase the water temperature decreased from 12.9 to 11.73 °C (on September 12th at 3:00). EC was positively correlated to the temperature, and decreased from 493 to 438 μS/cm (on September 12th at 4:00).

After this rain event the water level started to decrease, whereas water temperature and EC continuously increased.

This interval presents the conditions of low waters. After rain event allogenic recharge from the Manastirečka Reka river was activated which flows quickly through the karst system. One day after the rain, water level rises sharply, whereas EC and temperature dropped quickly as a result of the fast circulation of low mineralized water within the system.

Time interval between March 7th and March 17th, 2016

In the period between February 28th and March 7th 2016 only 2.5 mm of rain had fallen in the study area. Water level varied between 68.54 and 73.51 cm, temperature oscillated between 9.61 and 9.68 °C, and EC was in interval between 246 and 250 μS/cm.

The rain on March 8th (19.5 mm) caused rise of the water level to 76.38 cm, and sharp decrease of temperature (9.4 °C) and EC (228 μS/cm). The rain that followed on March 10th (13 mm) caused continuous increase of water level (76.14 cm) and simultaneous decrease of temperature (9.46) and EC (234 μS/cm) at the same day at 21:00. Very little rains on March 11th and 12th (2 mm and 0.5 mm, respectively) caused a small increase of water level, and small decrease of temperature and EC. More intensive rains on March 13th and 14th (12.5 mm and 7 mm, respectively) caused continuous rise of water level. Maximal value was reached on March 14th at 11:00 (78.80 cm). The water temperature decreased and reached the lowest value of 9.33 °C in the early morning on March 15th. EC also decreased during the whole day on March 14th when it reached value of 222 μS/cm. Low rain on March 16th (3 mm) did not cause significant changes to the physical characteristics of the spring water (Figure 5).

After this rain period, the rain stopped and the water level started continuously to decrease, whereas water temperature and electrical conductivity simultaneously increased.

The interval between March 7th and March 17th 2016 presents the conditions of high waters when the pores of the karst aquifer are filled with water and are hydraulically connected. Rain events in the recharge area of the Manastirec spring cause rapid reactions in the system. The water level increases very fast, on the opposite, the water temperature and electrical conductivity dropped quickly because of the low mineralization of the ground water and fast crossing the karst system toward the spring.

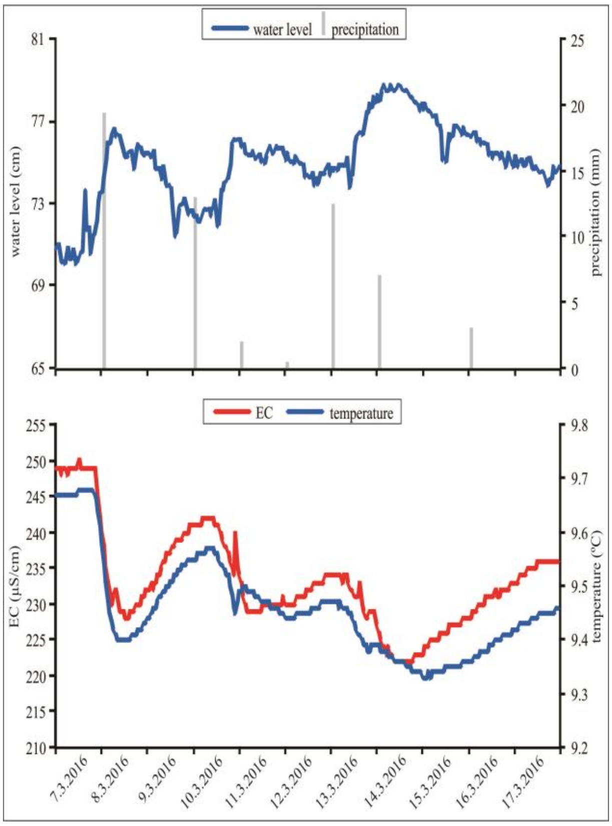

Fig. 5. Measurements of precipitation, water level, temperature and electrical conductivity (EC) in the period between March 7th and March 17th, 2016, every hour

Time interval between September 5th and September 14th, 2016

During the August 2016, in the study area, 59.5 mm of rain had fallen. In the period between August 28th and September 5th 2016 there was light rain (4.2 mm). In this period water level varied between 44.01 and 49.98 cm, water temperature was in interval between 11.2 and 11.57 °C, and EC oscillated between 374 and 413 μS/cm.

On September 6th there was light rain (1.2 mm). More intensive rains occurred on September 7th (53.2 mm) and September 8th (57.9 mm) and caused increase of water level from approximately 46 cm (September 6th) to 61.02 cm on September 8th at 17:00 (Figure 6). At this time, the water temperature rapidly decreased from 11.59 °C (September 6th) to 11.01 °C (in midday on September 8th), and EC showed the same oscillations as temperature and values dropped from 414 (September 7th) to 334 μS/cm (on September 8th at 20:00). The rains in the next five days (total amount of 17.9 mm) caused slow increase of water level and slow decrease of the temperature and EC (Figure 6).

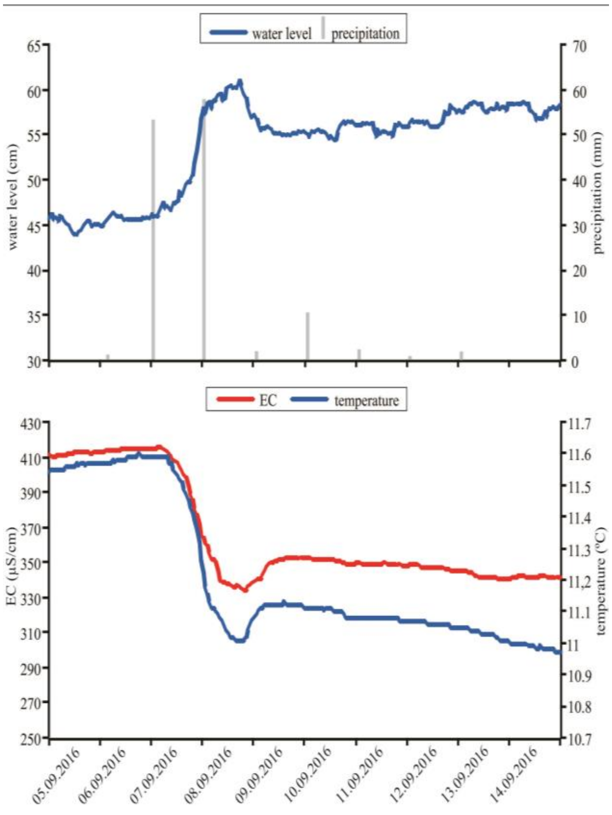

Fig. 6. Measurements of precipitation, water level, temperature and electric conductivity (EC) in the period between September 5th and September 14th, 2016, every hour

The analyzed period shows the conditions of the karst system during low waters. Fast changes of water level, water temperature and conductivity after rain event show that the Manastirec spring is characterized by fast reactions to hydrological events. The sharp increase of the water level which is followed with decrease of EC is due to dilution of pre-event water with water from the rain. The infiltrated waters into the karst system have low level of mineralization which is the reason why electrical conductivity drops sharply after the rains. The values of water temperature show the positive correlation to the conductivity which indicates inflow from surface waters.

Dynamics of hydrochemical parameters during the rain events in July and August, 2016

During the June 2016, in the area, total of 16.7 mm rain had fallen. In the period between July 1st and July 15th, 2016, 41 mm of rain had fallen with maximum on July 3th (36 mm). During the period between July 1st and July 15th, the water level varied from 49.56 to 60.15 cm, the water temperature oscillated between 10.66 and 11.12 °C, and EC was between 321 and 389 µS/cm.

The rains on July 16th and July 17th (25.5 mm and 0.4 mm, respectively) caused slow increase of water level from 50.64 cm to 53.74 cm. At the same time, temperature sharply decreased from 11.13 to 10.97 °C, and EC dropped from 374 to 348 µS/cm (Figure 7). In this time interval the sulphate increased from 33.41 to 47.98 mg/l, Ca/Mg rate increased from to 2.3 to 2.9, while nitrate decreased from 3.09 to 1.32 mg/l. Also, calcium level increased (up 4.46 meq/l), while magnesium level was slightly increased (1.57 meq/l). Rise in Ca/Mg rate with simultaneous drop in EC and water temperature, and nitrate reflects the impact increase of the fast flowing freshly infiltrated water in the recharge area of the Manastirec spring. According to the low Ca/Mg ratio we would infer that an important inflow are the ponors in Manastirečka Reka which are built in dolomite marbles.

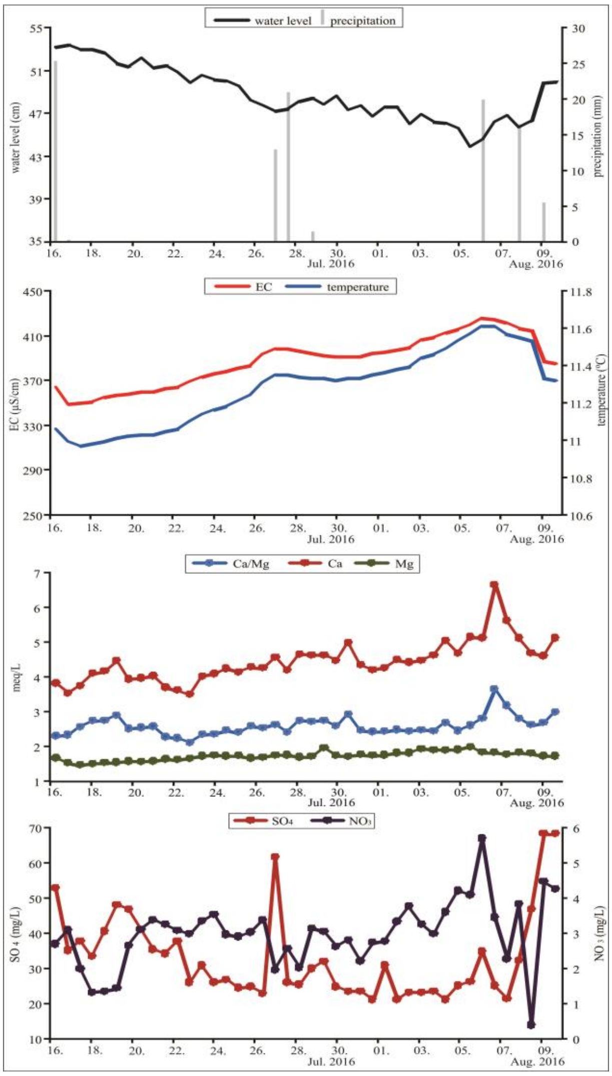

Fig. 7. Variations of water level, electrical conductivity (EC), water temperature and some hydrochemical parameters at the Manastirec spring

In the period between July 18th and July 25th there was no rain event, and water level continuously decreased to 47.09 cm, while water temperature values expressly rose to 11.32 °C, and EC increased to 394 µS/cm. In this dry interval the sulphate decreased to 22.84 mg/l, nitrate increased from 2.65 to 3.38 mg/l, and Ca/Mg ratio showed variations between 2.11 and 2.58.

More intensive rains that followed on July 26th, 27th and 28th (total amount of 35.5 mm) caused a small increase of water level (48.59 cm), and decrease of the temperature (11.32 °C) and EC values (389 µS/cm). In this interval the concentration of sulphate sharply increased to 61.69 mg/l, the Ca/Mg ratio showed small increase (2.92), and the nitrate decreased to 1.96 mg/l. All above mentioned parameters point out a dilution effect which indicates the rapid arrival of infiltrated rainwater throw the system in the spring.

The next eight days were without rainfall. In this period water level showed permanent decrease. On the other hand, temperature and EC showed continuous increase. During the whole analyzed period, the minimal water level of 43.43 cm was recorded in the afternoon on August 6th. The water temperature reached the maximal value of 11.65 °C also on August 6th, while EC increased to 430 µS/cm (August 7th). During the discharge decrease, sulphate slowly decreased, nitrate rose and reached the maximum value (5.69 mg/l) on August 6th, whereas Ca/Mg ratio showed uniform values which varied between 2.42 and 2.60.

After reaching the minimal discharge of the Manastirec spring very intensive rains on August 6th (20 mm), August 8th (16.5 mm), and August 9th (5.5 mm) caused discharge increase. The highest value of the water level (52.32 cm) was recorded in the early morning on August 11th, while temperature and EC continuously decreased and reached the minimal values in the morning on August 12th (11.15 °C and 367 µS/cm, respectively) (this is not shown on Figure 6). After this rainy interval, dry days followed, which caused decrease of water level and increase of both temperature and EC. In the interval between August 6th and August 9th sulphate sharply increased to 68.27 mg/l and Ca/Mg ratio rose to 2.98, and maximal values were reached on August 9th. The nitrate sharply decreased to 0.38 mg/l on August 8th. At this time we recorded 4.61 meq/l calcium and 1.72 meq/l magnesium. A fast increase of water level which was followed with decreasing of EC, water temperature and nitrate and increasing of sulphate and Ca/Mg ratio is due to dilution of the pre-event water with water from the rain events, and inflow from the river of Manastirečka Reka.

CONCLUSION

We observed the variability of the different hydrological parameters during several rain events in 2015 and 2016. Researches of the karst spring Manastirec showed that natural tracers as rains, temperature, electrical conductivity and water level can be better choice than the more complicated artificial tracing tests. Also, sulphate and nitrate levels, as well as the Ca/Mg ratio could be an important natural tracer within the multiparameter monitoring of transport of contaminants through the system. Monitoring of these parameters indicated the properties of the ground water flow and transport of contaminants from a recharge area of the spring.

The Manastirec spring shows a high temporal variability of hydrologic, physical and chemical parameters. Spring hydrographs respond rapidly and strongly to rainfall events which show that infiltrated rains into vadose zone flows quickly toward the spring. About one day after the rains, water level rises sharply, whereas both, electrical conductivity and water temperature fall quickly. The electrical conductivity values react fast as a result of the rapid circulation of low mineralized, newly infiltrated water within the karst system. The water temperature value is positively correlated to the electrical conductivity which indicates inflow from surface waters. Our research points out that rain events and chemical responses are controlled by dilution and water-rock interactions. Also, the results show fast arrival of the soluble substances from the surface through the system, which indicates high vulnerability of the Manastirec spring to contamination from the Manastirečka Reka river.

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Р е з и м е

ПРИРОДНИ ТРАСЕРИ КАКО АЛАТКА ЗА РАЗБИРАЊЕ НА ФУНКЦИОНИРАЊЕТО НА КАРСТЕН СИСТЕМ И НЕГОВА РАНЛИВОСТ
– АНАЛИТИЧКА СТУДИЈА ЗА ИЗВОРОТ МАНАСТИРЕЦ –

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Клучни зборови: природни трасери; хидрохемија; карстен извор Манастирец

За карсниот извор Манастирец лоциран во долината на Манастиречка Река, Порече, се анализирани различни хидролошки параметри за време на неколку дождовни периоди во 2015 и 2016 година. Истражувањата на извор-

ската вода покажаа дека користењето на природните трасери, како што се: врнежите, температурата на вода, електричната спроводливост и нивото на вода, претставува соодветен избор во споредба со сложените вештачки трасери. Исто така, нивото на сулфати и нитрати, како и односот Ca/Mg, се значајни природни трасери при мониторингот на транспортот на контаминанти низ карсниот систем. Следењето на овие параметри се покажа како добар показател за динамиката на подземните води и транспортот на загадувачите од водоносната зона изворот. Изворот Манастирец покажува големи временски варијации на хидролошките, физичките и хемиските параметри на водата. Преку хидрографот се гледа брзо и изразено движење на инфилтрираните врнежи преку воадоносната зона до изворот. Околу еден ден по изразени врнежи нивото на водата брзо расте, додека електричната спроводливост и температурата на водата брзо опаѓаат. Вредностите на спроводливоста покажуваат брза реакција поради рапидната циркулација на нова и слабо минерализирана вода инфилтрирана во карсниот систем. Вредностите за температура на водата се во корелација со електричната спроводливост, што укажува на прилив на вода од површината на теренот. Истражувањата покажаа дека разредувањето и интеракцијата вода–карпа се контролирачки процеси. Брзото движење на растворените материи од површината на теренот низ карсниот систем прават изворот Манастирец да биде силно ранлив од контаминантите кои доаѓаат од Манастиречка Река.