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A STABLE ISOTOPE INVESTIGATION OF THE CLASSICAL KARST AQUIFER: EVALUATING KARST GROUNDWATER COMPONENTS FOR WATER QUALITY PRESERVATION

RAZISKAVA KRAŠKEGA VODONOSNIKA S STABILNIMI IZOTOPI

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Daniel H. Doctor & Sonja Lojen & Milena Horvat: Raziskava kraškega vodonosnika s stabilnimi izotopi

Vodonosnik Krasa napaja voda različnega izvora. Namen študije je bil ugotoviti, kakšen je vpliv posameznih komponent v spremenljivih hidroloških pogojih na izvire v vznožju in obrobju kraške planote. Kot naravna sledila so bili uporabljeni stabilni izotopi in živo srebro, katerega koncentracije so v reki Soči značilno povišane.Rezultati izotopskih meritev so potrdili razdelitev kraške podtalnice v 3 kategorije: (1) izviri in estavele ob presihajočem jezeru (Sabliči, severni izvir Moščenice, Doberdobsko jezero), (2) skupina izvirov Timave, ki so pod močnim vplivom reke Reke in infiltracije padavin, in (3) izviri Sardoča in južni izvir Moščenice, ki predstavljajo vmesno kategorijo med prvima dvema. Koncentracije živega srebra so v splošnem nizke, vendar značilno odvisne od hidroloških pogojev, kar kaže na spremenljiv vpliv reke Soče.
Ključne besede: kras, podtalnica, stabilni izotopi, živo srebro, Kras, Slovenija, Italija.

Abstract UDC: 556.33

Daniel H. Doctor & Sonja Lojen & Milena Horvat: A stable isotope investigation of the Classical Karst aquifer: evaluating karst groundwater components for water quality preservation

The karst aquifer resurgence zone that is located along the western border of the Classical Karst region of southwestern Slovenia and to the north of Trieste, Italy is comprised of several distinct groundwater components. The purpose of this ongoing study is to examine the varying influence of these groundwater components on the karstic outflow under changing hydrologic conditions, using natural stable isotopes as tracers. In particular, the influence of the Soča river on the groundwater of this region was examined using mercury, a pollutant of elevated concentration in the Soča, as an additional tracer of Soča river water.The results of the isotopic measurements confirm the division of karstic groundwaters into three main categories: (1) springs and the estavelles of an ephemeral karstic lake (Sablici springs, Moschenizze North spring, Doberdò Lake), which are largely influenced by the Soča and Vipava rivers during periods of low flow, (2) the group of the Timavo springs that is subject to main influences of the Reka River and water derived from local precipitation, and (3) the grouping of Sardos spring and Moschenizze South spring, which form an intermediate category between the first two groups, exhibiting characteristics that indicate variable contributions from the other two end-members. Mercury levels in these karstic groundwaters are generally quite low, however significant variablity in mercury levels with varying hydrologic conditions have been observed, indicating also a varying influence of the Soča river.
Key words: karst, groundwater, stable isotopes, mercury, Kras, Slovenia, Italy.

INTRODUCTION

The Classical Karst region, located in southwestern Slovenia and stretching along the coast of the Gulf of Trieste across the southern border between Italy and Slovenia, is an area well known for its karstic features. The first geographical study of any karstic terrain was conducted in this region, the area being systematically described for the first time as early as the 17th century. Despite its distinguished place in the history of karst science, relatively little is yet known of the hydrogeology of this region. Over the course of the last century relatively few hydrogeologic investigations have been conducted in this region, and these have been mainly limited to the results of tracer experiments. Given that this is a water-scarce region, which is nonetheless home to several thousands of people, more detailed studies of the hydrogeology of the area are warranted.

Geologically, the Karst region is a large anticlinal block of Mesozoic limestones overthrust upon younger Eocene-age sandstones and shales, and forming an elevated plateau of nearly 400 square kilometers adjacent to the Adriatic Sea. The surface of this plateau is a mature karst terrain, characterized by numerous dolines and caves, deep shafts, and a conspicuous lack of surface water. Though the region receives, on average, over 1200 mm of rainfall each year, this water rapidly infiltrates into the bedrock and is not retained on the surface. Where accessible, the water table is very deep, reaching depths of over 300 m in some places. As a result, the inhabitants of the Karst region have looked toward the southwestern edge of this area for a source of fresh water, where the resurgence zone of the underground karstic aquifer exists. Water from this zone supplies the inhabitants of the Karst region in Slovenia, as well as partially supplying the water for the city of Trieste in Italy.

The karst aquifer is supplied by several distinct water components. These include: 1) groundwater derived from local meteoric precipitation on the Karst area, 2) allogenic water derived from sinking rivers and streams (most notably the Soča and Vipava rivers along the eastern border of the Karst) and 3) allogenic groundwater derived from the underground flow of the upper Timavo (Reka) river. These separate components mix within the karst aquifer and are discharged at the resurgence of the aquifer system, which is made up of many different springs. Due to the establishment of a comprehensive hydrologic monitoring network and the results of several tracer investigations, much is known regarding the basic hydrological characteristics of this system. The state of the present knowledge was well summarized by Civita et al. (1995). Despite the information that already exists, key questions remain concerning the origin of the water that is discharged from the springs in the aquifer resurgence zone, particularly regarding the influence of surface rivers on the groundwater of this region. Prior research has shown that both the Vipava and Soča Rivers contribute to the groundwater of this aquifer, however estimates of changing seasonal contributions from these rivers have not yet been established.

The purpose of this ongoing study is twofold: 1) to examine the varying influence of the different groundwater components on the karstic outflow under changing seasonal hydrologic conditions using stable isotopic measurements, and 2) to further examine the influence of the Soča River on the groundwater of this region using both stable isotopes and mercury, a pollutant of elevated concentration in the Soča, as an additional tracer of Soča river water.

METHODS AND RESULTS

Samples of spring, river, and well waters were taken each month for a period of nearly one year. The sampling locations are shown in Fig. 1. Water samples were collected for the analysis of the hydrogen stable isotopic composition of the water (δ^2H), and for the carbon stable isotopic composition in dissolved inorganic carbon ($\delta^{13}C_{DIC}$). Though normally reported in the literature, the oxygen isotopic values of these waters are not reported here; the separations between the $\delta^{18}O$ values of these waters are often less than twice the standard deviation of the measurements, thus making the oxygen isotope measurements difficult to interpret. Selected samples were collected for analysis of total and dissolved mercury. All of the isotopic data are presented in Fig. 2 and Fig. 3 in time series, from November 1998 to October 1999. From these charts, several conclusions can be made:

First, the long-term hydrogen and carbon isotopic compositions of the karstic waters exhibit opposing trends to one another. While the δ^2H values of all spring waters exhibit a progressive negativization throughout the summer followed by an abrupt increase toward more positive values in mid-October, the $\delta^{13}C$ values show a progressive increase during the summer, and rapid negativization in October.

Regarding hydrogen, one might expect the karst groundwaters to exhibit more positive isotopic values in the warmer summer months and more negative values during the winter, thus following the seasonal isotopic trend of meteoric precipitation. Instead, the observed trend is an inverted one, in which more negative values occur during the warmest months, and more positive values during the coldest months. This strong isotopic inversion was first observed in these springs by Flora and Longinelli (1989), who concluded that the aquifer discharge rises from at least two-possibly three-different reservoirs stored within the karstified bedrock. They assert that the first reservoir is one of more isotopically depleted water that gains its recharge from the internal, eastern Karst area at a mean elevation of 800 to 900 meters a.s.l., while the second is a more isotopically enriched reservoir that gains its recharge from local precipitation falling along the coast and on the inland plateau at a mean elevation of 300 to 400 meters a.s.l. The third reservoir they suggest is one fed directly by infiltration of Soča (Isonzo) River water into the karstic aquifer. This third reservoir is believed to influence the spring dicharge only during extended periods of low rainfall, and to yield the most negative isotopic signatures of the springs. Due to the high-altitude drainage basin of the Soča River, the river's recharge is much more isotopically depleted than the precipitation falling on the littoral Karst, particularly during the spring season when melting snow in the Slovenian mountains greatly contributes to the river's discharge.

While the hydrogen data presented here support the conclusion that the springs are recharged by different reservoirs of water under different hydrologic conditions, the origins of these reservoirs deserve closer inspection. Here is where the carbon isotopes, which have not been looked at previously, are most useful. The carbon isotope data show that the most negative values of dissolved inorganic carbon (DIC) in the spring discharge occur during the wettest periods of the year, namely during the early spring and fall. In the early spring of 1999, after a snowmelt event, there occurred a pronounced negativization in the carbon isotopic values of all of the spring waters. This observation is consistent with the so-called *piston-flow model* of karst circulation in which water held in storage in the slower-circulating part of the aquifer is flushed through minor fractures toward the larger conduits of the main drainage, with little mixing occurring (Ford and Williams, 1989).

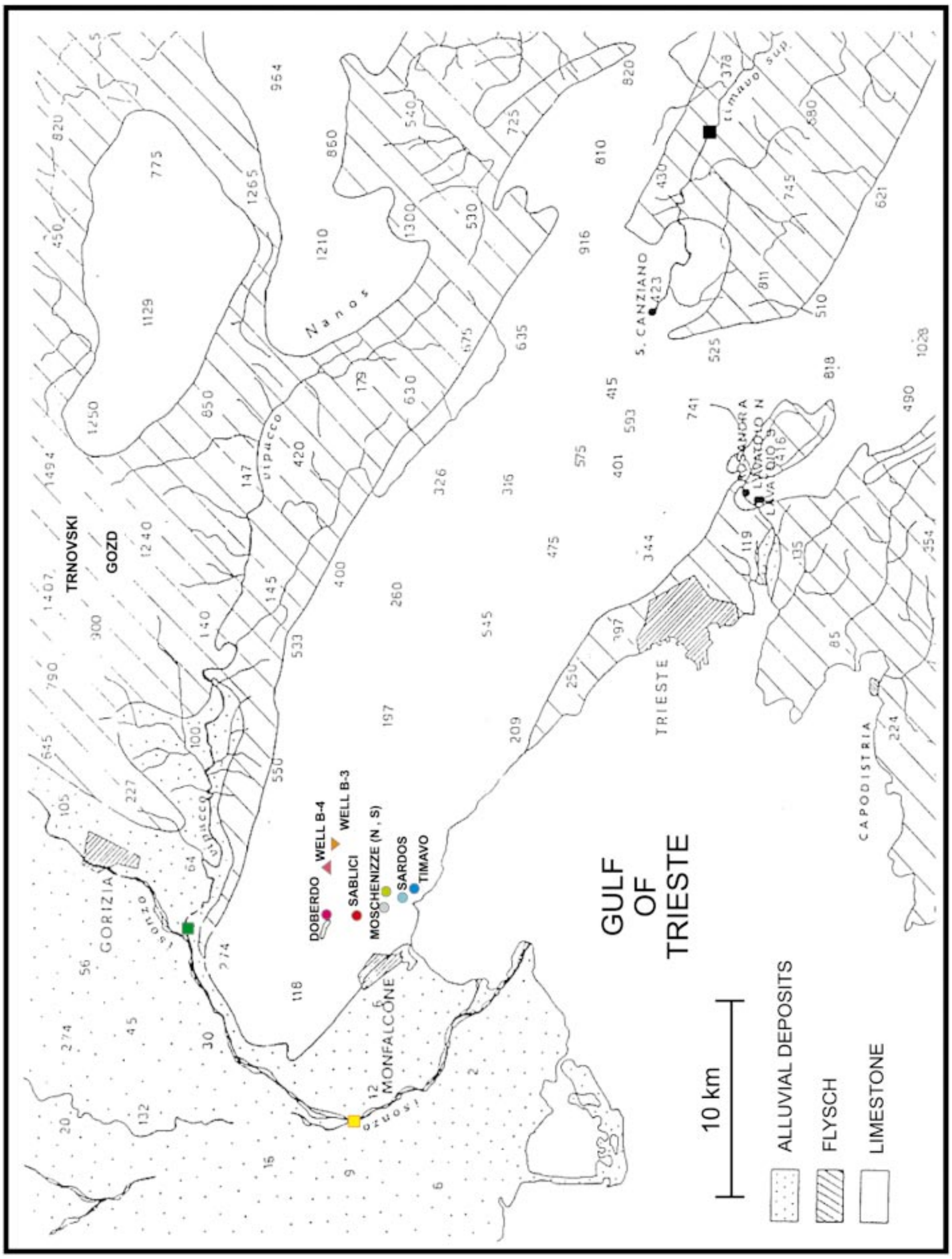

Fig. 1: Map of Karst region with sampling stations. The small numbers indicate surface elevation above sea level in meters (after Flora and Longinelli, 1989).
Sl. 1: Karta Krasa z zajemnimi mesti. Številke so nadmorske višine površja v metrih (po Flora in Longinelli, 1989).

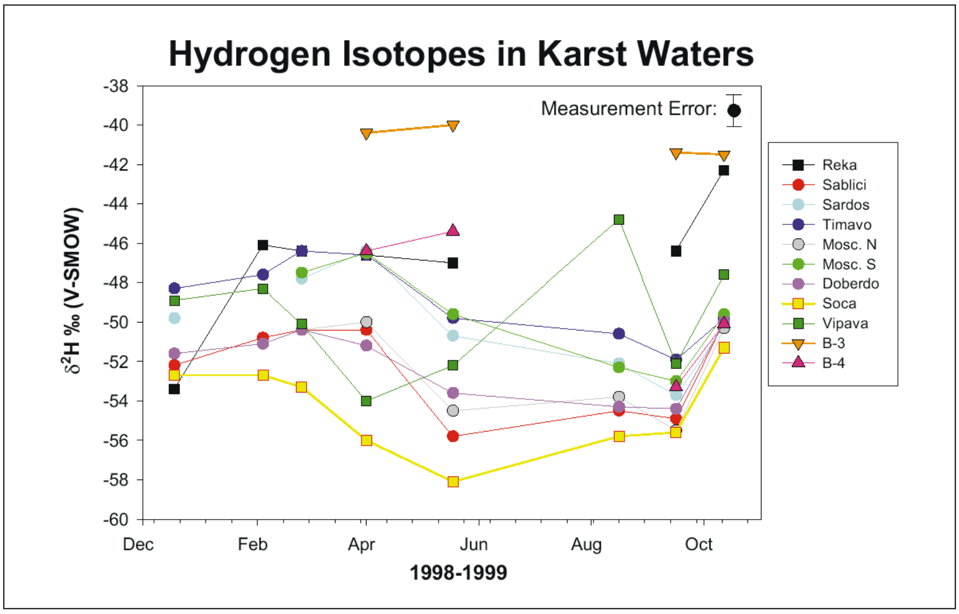

Fig. 2: Hydrogen isotopes in karst waters.
Sl. 2: Izotopska sestava vodika v kraških vodah.

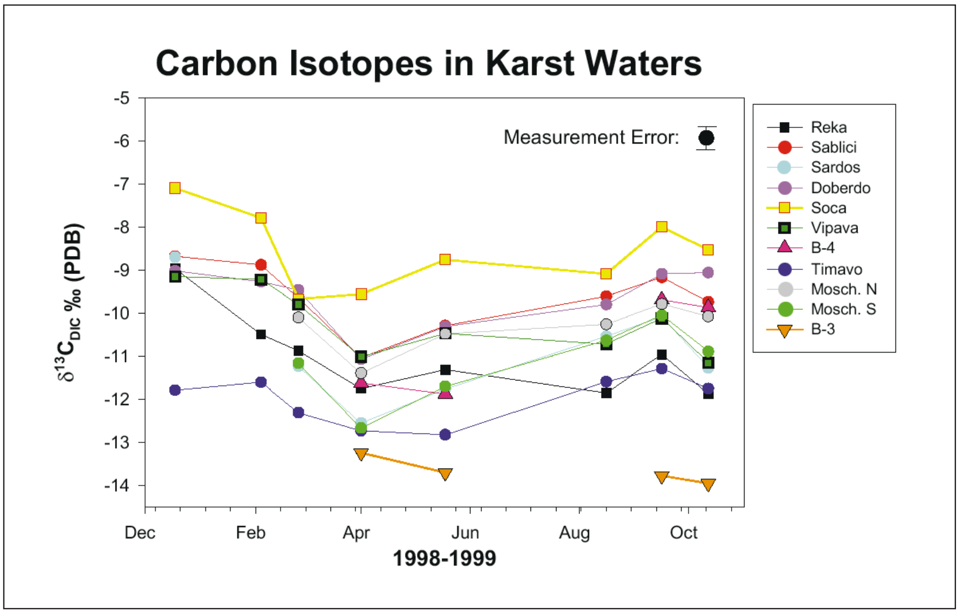

Fig. 3: Carbon isotopes in karst waters.
Sl. 3: Izotopska sestava raztopljenega anorganskega ogljika v kraških vodah.

Water stored within the unsaturated zone is subject to open-system equilibrium conditions, therefore the isotopic composition of the DIC in the water reflects that of a mixture of dissolved carbonate obtained form the host limestone rock and that of dissolved soil CO_2. Meteoric precipitation, which gains all its DIC from atmospheric CO_2, generally has $\delta^{13}C_{DIC}$ values around -7.0 ‰, while waters in isotopic equilibrium with the carbonate bedrock have $\delta^{13}C_{DIC}$ values around 0.0 ‰. An average $\delta^{13}C$ value for soil CO_2 in the Karst region of Slovenia is approximately -21.0 ‰ PDB, and unsaturated zone waters measured within a cave in the region showed average $\delta^{13}C_{DIC}$ values in the range of -7.0 ‰ to -16.0 ‰ (Vokal *et al.*, 1998). Similarly, the range of $\delta^{13}C_{DIC}$ values of the spring waters is -8.6 ‰ to -12.7 ‰, thus indicating a variable influence of soil CO_2. The waters having the lightest $\delta^{13}C_{DIC}$ values are the most heavily influenced by soil CO_2, and have thus been stored within the unsaturated zone for a longer period of time in order to obtain such light $\delta^{13}C$ isotopic signatures.

Given that the lighter carbon isotopic signatures coincide with heavier hydrogen isotopic signatures, the unsaturated zone is the most probable location of the reservoir of water which contributes to the spring discharge during conditions of elevated hydraulic pressure in the aquifer (for example, after snowmelt or storm events). This reservoir is most likely replenished by local meteoric precipitation falling on the littoral Karst, in accordance with the model proposed by Flora and Longinelli, thus giving rise to the more isotopically enriched δ^2H values. During heavy storms or snowmelt events, the water held in storage in the unsaturated zone is flushed through the system as new water is added to the surface. The previously stored water then flows through the main drainage network and is ultimately manifested at the springs. During drier periods, the water that flows from the springs as baseflow most probably receives its recharge from a reservoir largely influenced by the Soča and Vipava rivers.

Some evidence of the widespread influence of Soča river water on the aquifer are the data from a pumping well at Klariči, hereafter referred to as well B-4. This well provides the water supply for the Slovene inhabitants on the Karst platueau. Urbanc and Kristan (1998) have reported on the results obtained from a prolonged intensive pumping test at well B-4, during which water samples were collected for oxygen isotopic analysis as well as for other chemical parameters. Their results show that as the piezometric level of the aquifer dropped during pumping, the isotopic and chemical composition of the water progressively shifted toward that of the Soča River along a mixing line between the river water and the local aquifer composition. Urbanc and Kristan estimated that the water drawn from the well during the pumping test contained between 50 % and 60 % Soča River water. Pezdič *et al.*, (1986) have also reported a similar proportion of Soča River water in this well.

The isotopic data presented here also support these observations. In September and October 1999, the hydrogen isotopic composition of nearly all of the karstic groundwaters strongly shifted toward the isotopic composition of the Soča river (see Fig. 2). The most dramatic shift occurred at the pumping well B-4, where the δ^2H composition of the water changed from -45.4 ‰ in May to -53.3 ‰ in September. During the summer, very little precipitation fell on the Karst region, thus causing a lowering of the piezometric level of the karst aquifer and allowing a greater influx of Soča River water. Taking the δ^2H stable isotope compostion of borehole B-3 to be representative of the local groundwater, the mixing proportion of Soča River water in the pumping well B-4 can be estimated for both the spring (April-May) and fall (September-October) seasons. In the spring, the proportion of Soča River water in the pumping well is estimated to have been 34%. During the fall, the percentage of Soča River water in the pumping well increased dramatically, to an estimated 86%.

A similar shift can also be observed in the carbon isotopic data, where the $\delta^{13}C_{DIC}$ value of the well water changed from -11.88 ‰ to -9.69 ‰ over the same time period, from spring to fall of 1999. It is interesting to note that the carbon isotopic values of the other groundwaters did not experience the same degree of shift as the well B-4.

CLASSIFICATION OF THE KARSTIC WATERS

The results of the isotopic measurements confirm the existence of a division between groundwater in this resurgence zone, as has been previously proposed by other authors (Gemiti & Licciardello, 1977; Gemiti, 1994; Civita *et al*., 1995). This divide effectively separates the rising karstic groundwaters into three main categories. The first is a grouping of several springs and the estavelles of an ephemeral karstic lake (Sablici springs, Moschenizze North spring, Doberdò Lake), which are likely influenced by the Soča and Vipava rivers during periods of low flow. The second is the group of the Timavo springs, which forms a category of its own subject to two main influences: the Reka river sinking at the Škocjan Caves in Slovenia, and water derived from local meteoric precipitation on the Karst region. The third is the grouping of Sardos spring and Moschenizze South spring, which form an intermediate category between the first two groups, exhibiting characteristics that indicate variable contributions from the other two end-members.

The following table summarizes the average values of the measured hydrochemical and isotopic characteristics of these three categories, as well as of the other waters studied:

Table 1: Average values of hydrochemical and isotopic measurements (each value represents the average of 11 samples taken monthly).
Tabela 1: Poprečni rezultati hidrokemičnih in izotopskih meritev (vsaka vrednost je poprečje 11 mesečnih vzorcev).

	$\delta^{13}C$ (‰ PDB)	**$\delta^{2}H$ (‰ V-SMOW)**	**Conductivity (μs/cm)**	**Temperature (°C)**
Doberdò	-9.63	-52.1	322	12.0
Sablici	-9.63	-52.4	320	12.1
Mosch. North	-10.35	-52.4	328	12.2
Sardos	-10.88	-50.1	379	12.6
Mosch. South	-11.18	-49.7	371	12.6
Timavo	-11.98	-48.9	387	11.9
Reka	-11.01	-46.9	306	8.8
Soča	-8.93	-55.0	268	12.3
Vipava	-10.20	-49.8	326	11.7
B-4/Klariči	-10.77	-48.8	490	14.0
B-3	-13.66	-40.8	494	13.4

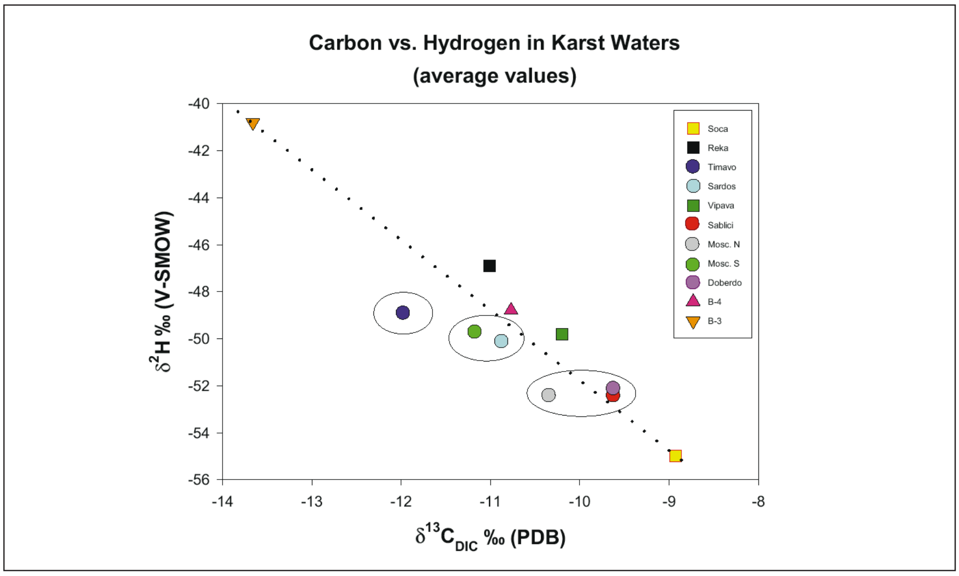

Fig. 4: Hydrogen vs. carbon isotopes (average values).
Sl. 4: Zveza med izotopsko sestavo vodika in raztopljenega anorganskega ogljika (povprečne vrednosti).

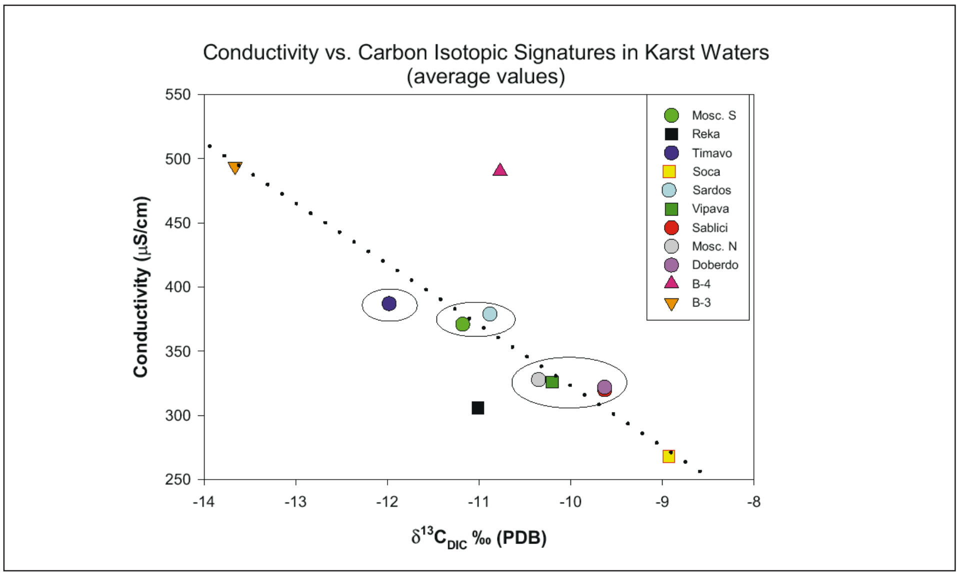

Fig. 5: Carbon isotopes vs. conductivity (average values).
Sl. 5: Zveza med prevodnostjo in izotopsko sestavo raztopljenega anorganskega ogljika (povprečne vrednosti).

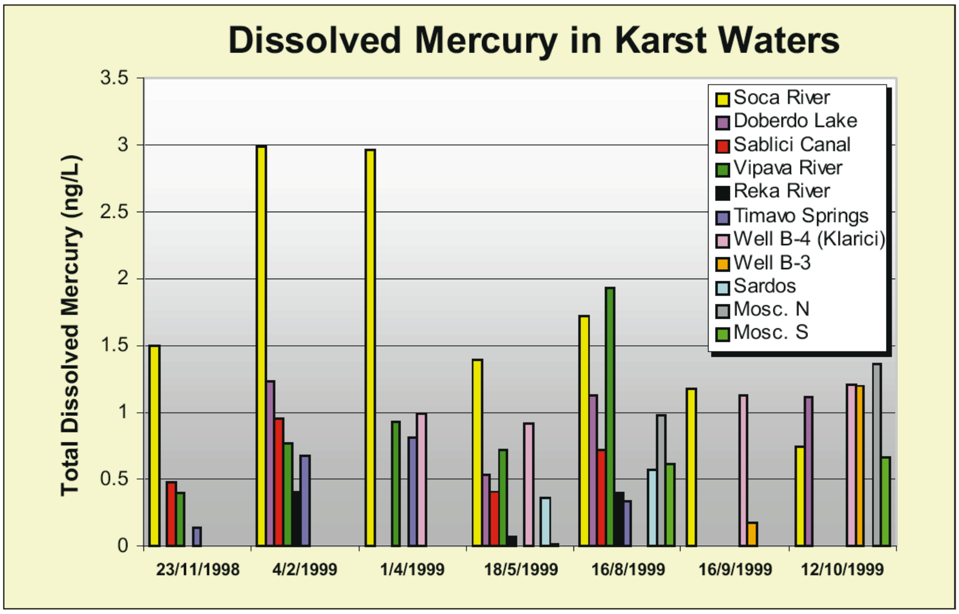

Fig. 6: Dissolved mercury in karst waters.
Sl. 6: Koncentracija celokupnega raztopljenega živega srebra v kraških vodah.

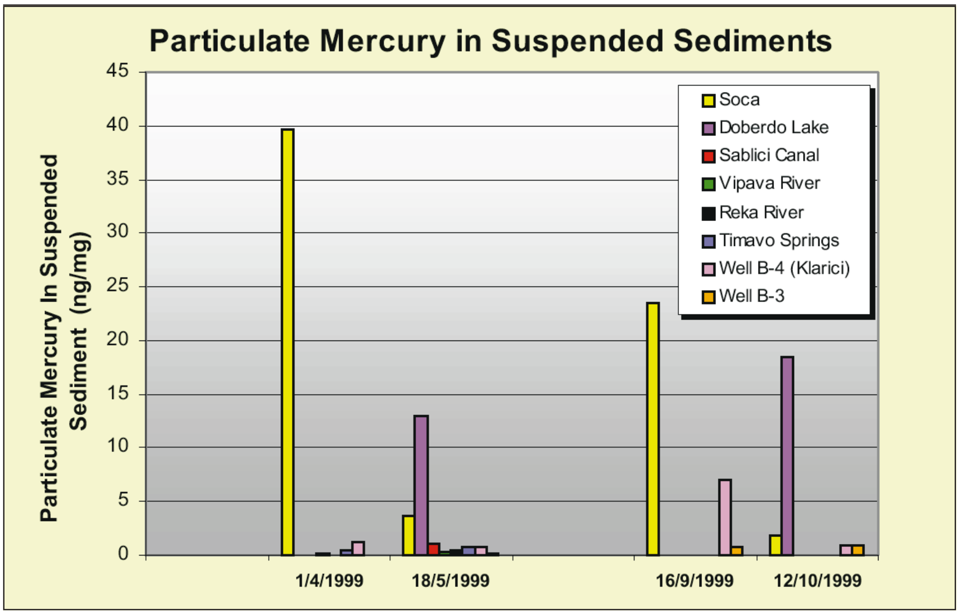

Fig. 7: Particulate mercury in karst water samples (calculated).
Sl. 7: Partikulatno živo srebro (izračunane vrednosti) v kraških vodah.

From these data, it is evident that the Doberdò/Sablici/Moschenizze North category exhibits the lowest conductivities, most depleted δ^2H signatures, and most enriched $\delta^{13}C$ signatures than the other spring waters. It is noteworthy that Moschenizze North spring has the most depleted $\delta^{13}C$ values and the highest conductivity water, indicating that this spring receives a greater proportion of its flow from water stored in the unsaturated zone than do the Doberdò and Sablici springs, the latter two being essentially identical to one another.

The Timavo Springs category exhibits the highest conductivity water, with the most enriched δ^2H signatures and most depleted $\delta^{13}C$ signatures of all the spring waters. Hydrochemical seasonal variability of the Timavo spring discharge is most definitely linked to the hydrochemical variability of the Reka River, as clearly demonstrated by simultaneous measurements in time series of conductivity, temperature and discharge at both sites (Cucchi *et al.*, 1997). However, the Timavo springs necessarily receive a great portion of their discharge from other sources, given that their average discharge is about twice the average discharge of the losses of the Reka River (Civita et al., 1995). The enriched hydrogen values are most likely the result of mixing between unsaturated zone water and Reka river water, the latter having the most enriched hydrogen signatures of all the three rivers sampled.

The Sardos/Moschenizze South category exhibits water with electrical conductivities, δ^2H signatures, and $\delta^{13}C$ signatures that are intermediate between the Doberdò/Sablici/Moschenizze North category and the Timavo Springs category. It is likely that the discharge of Sardos and Moschenizze South springs receive contributions from the other two reservoirs.

In Fig. 4, the carbon isotope signatures are plotted against the hydrogen isotopic signatures. From this chart it is evident that the average isotopic compositions of the spring waters lie along a mixing line between two end-members: the Soča River and borehole B-3. Given the hydrochemical and isotopic characteristics of the water sampled in borehole B-3 (see Table 1), we may consider boreole B-3 to be representative of the water in the inner karstic aquifer. The Soča River represents the other end of the chemical spectrum of the waters which feed the karst aquifer resurgence. Given the clear mixing relationship between these two main sources, we may conclude that all of the spring waters and the pumping well B-4 are affected, to some degree, by the Soča River. The three categories of the karstic resurgence proposed above are circled in the figure, and are clearly separate from each other. It is interesting to note that the Vipava River falls directly within the circle indicating the Doberdò/Sablici/Moschenizze North category. It is very likely that these springs receive a significant component of their discharge from the Vipava River, in addition to the Soča River. In fact, a direct connection between the Vipava River and the Doberdò/Sablici springs has already been established by artificial tracing (Timeus, 1928), so we may say with great certainty that the Vipava River also contributes to this spring category. However, given the relatively low gauged losses of the Vipava River (about 1.0 m^3/s) it is still uncertain how the contribution from the Vipava River changes on a seasonal basis.

Fig. 5 illustrates the nearly linear relationship between the average conductivity of the spring waters and their average carbon isotopic signatures. This relationship is consistent with the hypothesis already presented above, namely that the waters of more depleted carbon isotopic composition are those which receive a greater contribution of water stored within unsaturated zone. Although neither the conductivity of the water nor the carbon isotopic values give any definitive indication of the age of the water, it is reasonable to assume that the higher conductivity, more depleted carbon waters are not fed directly by present-day meteoric precipitation, nor by direct infiltration from the Soča River.

MERCURY IN THE KARSTIC WATERS

The sample measurements of total dissolved mercury are presented in Fig. 6, and the calculated levels of particulate mercury in the samples are shown in Fig. 7. The levels of total dissolved and particulate-bound mercury measured in these karst waters are generally quite low (<1.5 ng/L). The highest levels of dissolved and particulate-bound mercury have been observed in Doberdo, Sablici, and Moschennize North springs, in well B-4, and, on one occasion, borehole B-3. It is interesting to note that the borehole B-3, located approximately 1 km from the pumping well B-4, does not exhibit such consistently elevated mercury levels as B-4, and yields water of significantly different isotopic composition than that of B-4. It is probable that the karstic groundwater divide runs between these two wells, but that they are nonetheless hydraulically connected. The elevated mercury levels observed in these wells, particularly of particulate-bound mercury, further indicates an influence by the Soča river, as there exist no other significant sources of mercury which could affect the groundwater in the region.

While the mercury levels do not indicate a threat to the water quality of these groundwaters, they do indicate a certain degree of mercury accumulation in the aquifer, most probably associated with the sediments trapped in the kartic voids. Further work ought to focus on the degree of mercury accumulation, and assessment of its effects on the karstic ecosystem.

CONCLUSIONS AND RECOMMENDATIONS

1. A river influence is ubiquitous in the resurgence zone. The Soča river seems to have an influence on the groundwater which is not local, but is fairly widespread, and is manifested in the strong isotopic inversion observed in all of the sampled springs. This could also possibly be an influence of the Vipava river, which shows a similar isotopic inversion trend, and for which traced connections have already been established. This is still unclear, and a quantification of mixing between water components needs to be done. In general, the Soča River isotopic signatures are much lighter than the Vipava signatures. Unlike the model proposed by Flora and Longinelli (1989), however, the isotopic inversion recognized in the data from the area could in fact be ubiquitous from all allogenic recharge areas: including the watersheds of the Reka, Vipava, and Soča rivers.
2. Among the spring waters, there seem to exist at least three, possibly four distinct water categories: Doberdo/Sablici/Moschenizze North springs, Sardos/Moschenizze South springs, and the Timavo springs. This is in accordance with previous research.
3. The mercury concentrations in these waters are quite low in these waters, and do not represent a water quality threat to the aquifer. The highest levels of mercury have been observed in those springs which the isotope data indicate are most influenced by the Soča River. Therefore, the mercury serves as an additional tracer of Soča river water in these springs. The well B-4 exhibits high levels of mercury associated with suspended sediments, indicating a probable accumulation of mercury in the sediments of the karst aquifer.

Given that two large surface rivers, the Soča and the Vipava, contribute significantly to this regionally important aquifer system, it is advisable that concerted efforts are made to preserve the water quality of these rivers in order to prevent the future degradation of the karst aquifer. In addition, since the discharge of the springs also draws upon water held in storage within the unsaturated zone above the aquifer, any future changes in the land use of the Karst region will also have an impact on the water quality. These factors need to be kept in mind when planning for future development in the region.

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RAZISKAVA KRAŠKEGA VODONOSNIKA S STABILNIMI IZOTOPI

Povzetek

Območje napajana vodonosnika Krasa obsega območje vzdolž zahodne meje Kraške planote v SW Sloveniji in območje severno od Trsta v Italiji. Voda, ki se zadržuje v vodonosniku, izvira iz različnih območij: (1) podtalnica, ki se napaja z neposredno infiltracijo padavin na območju Krasa, (2) alogena podtalnica, ki izhaja iz podzemnega toka reke Reke (Timave) in (3) alogena voda iz ponikalnic, predvsem Soče in Vipave vzdolž vzhodne meje Krasa. Posamezne komponente se mešajo med seboj v zlivnem območju, na kateremje tudi več izvirov. Namen študije je bil preučiti vpliv posameznih komponent na izvire na območju Krasa v spremenljivih hidroloških razmerah z uporabo stabilnih izotopov kot naravnih sledil. Posebej nas je zanimal vpliv reke Soče zaradi prisotnosti živega srebra kot potencialnega onesnaževalca, katerega koncentracije so v Soči povišane, in je zato v tem primeru lahko služilo kod dodatno sledilo za vodo iz Soče. Rezultati izotopskih meritev so potrdili predpostavke avtorjev prejšnjih študij. Kraška podtalnica se deli v 3 kategorije. Prvo predstavlja skupina izvirov in estavel presihajočega jezera (izviri Sabliči, severni izvir Moščenice, Doberdobsko jezero), ki jih napajata predvsem Soča in Vipava v času nizkega toka. Drugo skupino predstavljajo izviri Timave, ki se napajajo z vodo reke Reke, ki ponika v Škocjanskih jamah, in lokalnimi padavinami. V tretjo skupino sodita izvira Sardoč in južni izvir Moščenice, ki v bistvu predstavljata vmesno skupino med zgornjima dvema, s spremenljivim deležem vode iz enega ali drugega vira.Vsebnost živega srebra v podtalnici je v splošnem nizka, vendar se glede na spremenljive hidrološke pogoje zelo spreminja. Najvišja vsebnost raztopljenega in partikulatnega živega srebra je bila ugotovljena v Doberdobskem jezeru in Sabličih, v vrtinah B-4 in B-3 na slovenski strani in v severnem izviru Moščenice. Tako izotopska sestava kot koncentracije Hg kažeta na pomemben doprinos reke Soče.