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Integrating hydrogeochemical and isotope data in studying regional groundwater flow systems in the Great Hungarian Plain

Teodora Szocs[a]*, Shaun Frape[b], Rhys Gwynne[b]

[a]*Geological and Geophysical Institute of Hungary, Stefánia út 14, 1143 Budapest, Hungary*
[b]*University of Waterloo, Earth and Environmental Sciences, 200 University Avenue W., Waterloo, N2L 3G1, ON, Canada*

Abstract

Environmental isotopes (δ ^{18}O, δ D, δ ^{13}C, ^{14}C, ^{87}Sr/^{86}Sr) combined with hydrogeochemical data have been used in studying regional groundwater flow systems in the Great Hungarian Plain part of the Pannonian Basin. Oxygen and deuterium isotope data show groundwater is of meteoric origin, with isotopic signatures indicative of petroleum producing units in the deeper part of the basin. Based on the carbon isotopes analysed on dissolved inorganic carbon, the groundwater residence times vary from a few thousand to tens of thousands of years. Strontium isotopic signatures show enrichment where carbonate dissolution is indicated by increasing bicarbonate values in Upper Miocene aquifers.

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Keywords: groundwater; water-rock interaction; isotope; Pannonian Basin; Great Hungarian Plain

1. Introduction

Groundwater is widely used around the world and with the increase of population and potential effects of climate change there is an increasing need to understand hydrogeological systems at all scales. In many cases the same aquifers are the source of different, often competing usage, including agriculture, drinking water supply, industry, balneology and geothermal energy production. Special attention has to be paid to transboundary aquifers which sometimes can reach or even exceed 1-2 M km^2 (eg. Nubian aquifer in Northern Africa, Guarani aquifer in Latin America) requiring bilateral or even multilateral international cooperation. While surface water management, based on good hydrological studies, is an established practice in many countries, there is still much to be improved in

* Corresponding author. Tel.: +36-1-251-4678.
E-mail address: szocs.teodora@mfgi.hu

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groundwater management. This can be achieved by a science-based understanding of groundwater flow systems. This study illustrates how hydrogeology in combination with hydrogeochemical and isotope data can improve the understanding of regional groundwater flow systems, the age and origin of the groundwater, demonstrating how integrated data can be a useful tool in groundwater management.

2. Hydrogeological – hydrogeochemical setting

The Great Hungarian Plain (GHP) is a regional geographic feature of about 52000 km^2, which covers the largest sub-basin of the Tertiary Pannonian. It is an extensively studied basin in Hungary, up to about 7000 meters thick, averaging 2-3000 m.

It developed in the Early-Middle Miocene during subduction along the Carpathian mountain range which resulted in a back-arc style extension [1], [2]. A large lake (Lake Pannon) formed and deepened until ca. 9.8 Ma. The Upper Miocene sediment successions which infilled the basin form the large, regional aquifer of the Pannonian Basin. The Nagyalföld and Zagyva Formations (referred as Miocene A in figures) form the upper part of the aquifer, while the Újfalu Formation (referred as Miocene B in figures) which is the main thermal aquifer is the lower regional water bearing unit. Quaternary sediments with various thicknesses form the main drinking water aquifers in the GHP.

Generally, large scale hydrogeological studies have been carried out since the first part of the 20th century, but the most relevant studies can be connected to the work of Erdélyi [3]. He studied the hydrodynamics of the Hungarian basin using a very advanced approach for his time. In his concept the importance of the whole hydrogeological system, even taking into consideration the areas outside the country border were studied. His groundwater flow concept set up in the 1960s which took account of the relevance of regional and intermediate groundwater flow systems and defined the recharge and discharge zones, is still valid. Based on Tóth's theory [4], hydrogeological numerical models followed by an evaluation of isotope data brought further possibilities in the understanding and interpretation of data. The work of Stute and Deák [5] was the first complex evaluation applying environmental isotopes, noble gases and hydraulics to deep groundwater circulation systems, along a Quaternary and a Pliocene flow system. Further detailed studies for example by Varsányi et al. [6] and [7] have been carried out on the different parts of the GHP.

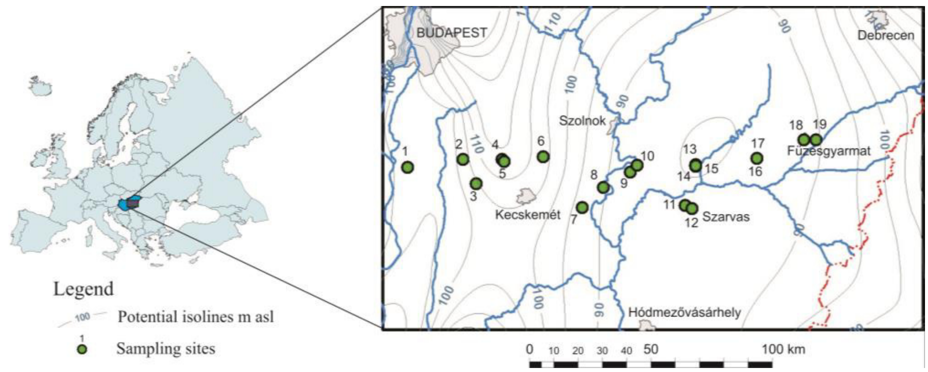

Fig. 1. Study area with sampling sites.

3. Groundwater characteristics

The hydraulic potential distribution (Fig. 1) modelled for groundwater in the upper part of the Újfalu Formation (thermal aquifer in the lower part of Upper Miocene) shows a minimum value of about 85 m asl around Mezőtúr (points 13, 14, 15), but the 90-95 m asl isolines can be considered a representation of the discharge zone both for the intermediate and the regional flow systems. A similar potential distribution (not shown in this paper) with about 2-5 m vertical differences was found for the upper Miocene Zagyva and Nagyalföld Formations and the Quaternary aquifers. The recharge zones of the study area can be found in the Danube-Tisza interfluve and outside the study area in the N-NE, E, and SE (the latter two located outside of the National border).

19 new groundwater samples (Fig. 1) were collected from operating wells during 2014 for analysis of major components, trace elements and environmental isotopes ($\delta^{18}O$, δD, $\delta^{13}C$, ^{14}C, $^{87}Sr/^{86}Sr$). The screen intervals vary between 60-330 m for Quaternary (points 1, 2, 4, 6, 8, 10), 160-860 m for Zagyva and Nagyalföld Formations (points 3, 11, 13, 14, 16, 18) and 300-1750 m for Újfalu Formation samples (points 5, 7, 9, 12, 15, 17, 19). The general hydrogeochemistry shows a $CaMgHCO_3$ water type for the Quaternary recharge areas, with an increasing Na content with depth. Where both local and intermediate flow systems are part of discharge areas the water chemistry is mainly a $NaMgHCO_3$ type. The deeper part of the intermediate and regional flow systems in the Upper Miocene formations are characterized by a $NaHCO_3$ water chemistry.

A strong correlation was found between the TDS and the depth distribution of the screen intervals of the studied wells. Groundwater based on samples used in the study, in the Quaternary sediments, show a low mineralization up to approximately 1000 mg/l. This value falls within the range shown by archive data. The Upper Miocene (Zagyva, Nagyalföld) formations are characterized by slightly higher TDS generally up to 2000 mg/l. The TDS in the main thermal aquifer (Újfalu Formation) studied varies between 500-5000 mg/l, which reflects good hydraulic conductivities and active flow systems.

The $\delta^{18}O$ and δD data of the sampled groundwater are shifted from the global meteoric waterline. With one exception (point 18) all the samples from the Quaternary and the upper Miocene Zagyva and Nagyalföld Formations lie on or near the local meteoric waterline with $\delta D = 7.9 * \delta^{18}O + 3.9$ (R^2=0.996) which is very similar to the meteoric waterline for the Carpathian basin $\delta D=7.8* \delta^{18}O+6$ defined by Deák [8]. The $\delta^{18}O$ data vary between -8.9 and -12.9‰, while the δD data range between -73.1 and -98.6‰ respectively.

$\delta^{18}O$ values of samples from the Újfalu Formation vary between -7.4 and -12 ‰. Some of the samples (e.g. 12, 15) are enriched in heavy oxygen isotopes. The enrichment in heavy oxygen isotopes most likely indicates mixing with formation waters associated with thermal waters from hydrocarbon bearing strata. Values of archive data from the lower Miocene formation waters are plotted on figure 2a (referred as Miocene C in figure).

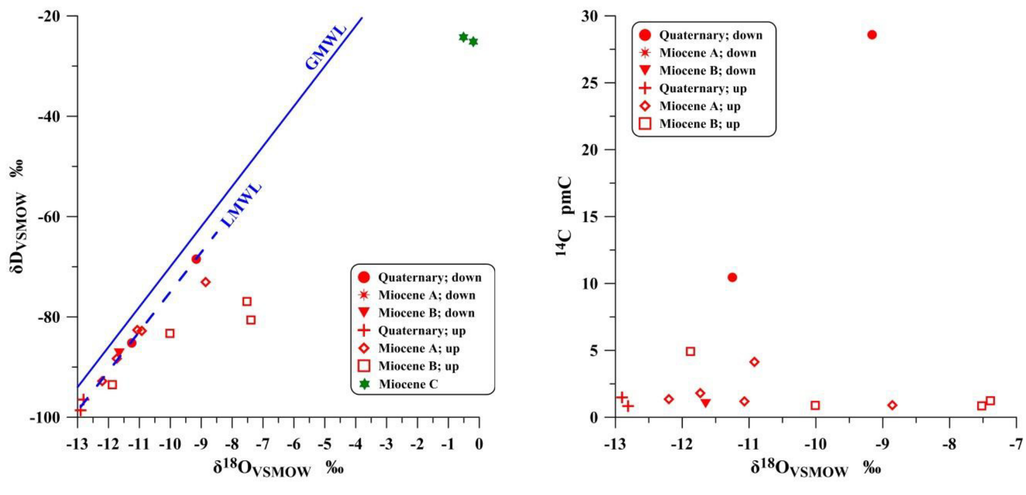

Fig. 2. (a) $\delta^{18}O$ versus δD (b) $\delta^{18}O$ versus ^{14}C

The ^{14}C data vary between near detection limit and ~29 pmC, with most of the samples having activities less than 2 pmC (Fig. 2b). These data are in accordance with the previous studies and hydraulic models and show that intermediate and regional groundwater flow paths both in the Nagyalföld, Zagyva and in the Újfalu Formations are on the scale of a few ten of thousand years or more. As with the applied ^{14}C method groundwater residence time determinations can be used up to about 40000 years, longer timeframe cannot be tracked. The most commonly used radiocarbon age calculation is based on the use of $\delta^{13}C$ values of DIC, which assumes that following the dissolution of CO_2 from the soil horizons, the DIC in groundwater originates from the dissolution of the aquifer's carbonates. The $\delta^{13}C$ values of the samples collected vary within a wide range from -27.9 to -0.8‰. Both bacterial and thermal degradation of organic matter can cause kinetic fractionation which in case of methanogenesis can be responsible for the carbon-13 enrichment of DIC. Similar enrichment was reported by Varsányi et al. [9] in the south eastern part of the Pannonian Basin and by Szocs et al. [10] in the western part of the Pannonian Basin.

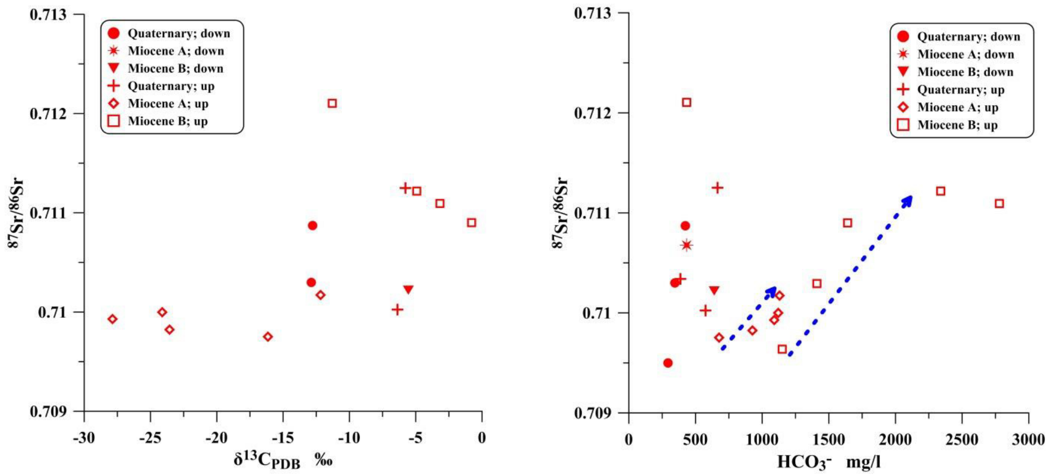

Fig. 3. (a) $^{87}Sr/^{86}Sr$ versus $\delta^{13}C$; (b) $^{87}Sr/^{86}Sr$ versus HCO_3^-: Blue arrows indicate carbonate dissolution trend

The $^{87}Sr/^{86}Sr$ ratios of groundwater are between 0.709 and 0.712 (Fig. 3). No correlation can be found between $^{87}Sr/^{86}Sr$ and $\delta^{13}C$ data (Fig. 3a) for groundwater from Quaternary and Nagyalföld, Zagyva Formations. In the latter case almost the same ratios were detected across the whole range of $\delta^{13}C$ values. An apparent negative correlation was found for groundwater in the Újfalu Formation, which is most likely due to the carbon-13 enrichment of DIC due to methanogenesis. A clear correlation can be detected between the $^{87}Sr/^{86}Sr$ and the HCO_3^- data (Fig. 3b) in the discharge zone for both Upper Miocene Formations, which shows carbonate dissolution has a role in the distribution of isotope ratios. Groundwater at Lakitelek with ratios above 0.712 must have a different recharge source than the other samples, which is supported by its very low TDS (~650 mg/l) at a depth of approximately 1000 m below surface. Cation exchange (on clay minerals) is probably also responsible for the increase of the $^{87}Sr/^{86}Sr$ ratio in groundwater in the Újfalu formation between Dévaványa and Mezőtúr.

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