HYDROGEOLOGICAL INVESTIGATION OF NATURALLY OCCURRING RADIONUCLIDES IN GROUNDWATER IN THE VICINITY OF THE VELENCE HILLS AND LAKE VELENCE

THESES OF PHD DISSERTATION

by

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1. Introduction and objectives

Groundwater is an important source of drinking water, but its use for drinking purposes may be limited by the excess of certain elements (e.g. As, Pb, U), which can be naturally enriched as a result of continuous rock-water interaction along the groundwater flow systems (EDMUNDS AND SMEDLEY 1996; VENGOSH et al. 2022).

Due to the continuous interaction with its environment, groundwater can contain naturally occurring alpha (^{238}U, ^{234}U, ^{232}Th, ^{226}Ra, ^{222}Rn and ^{210}Po) and beta-decaying radionuclides (^{40}K, ^{228}Ra and ^{210}Pb), from concentrations below the detection limit to concentrations that can threaten human health (HOEHN 1998; NUCCETELLI et al. 2012).

The continuous and organised flow of groundwater affects the spatial distribution of radionuclide concentrations in the groundwater (SKEPPSTRÖM AND OLOFSSON 2007; TÓTH 2009). In order to understand the processes affecting the quality of drinking water derived from groundwater resources, it is therefore essential to apply the groundwater flow system approach, which can be an effective tool in understanding water quality problems, thus contributing to the maintenance of a safe drinking water supply.

In my doctoral work, I conducted research in the surroundings of the Velence Hills and Lake Velence and aimed to evaluate the radionuclide content of groundwater in a groundwater flow system context to explore the spatial relationship between the type of groundwater flow systems and the radionuclide concentrations. To achieve my aims, I formulated the main objectives of the doctoral thesis as follows:

1. Investigating the properties of groundwater flow systems (driving force, regional flow directions, hydrochemical characteristics).

2. Explaining the spatial distribution of the measured radionuclide concentrations based on the revealed groundwater flow systems (regime types, relative residence time).
3. Integrating Lake Velence, a soda lake of social and ecological importance, into the groundwater flow systems, using the measured radionuclide concentrations as environmental tracers.
4. Understanding the geochemical processes controlling the mobilization of uranium in the Pannonian siliciclastic aquifer, as well as drawing general conclusions about the behavior of uranium in rock-water systems.

2. Applied methods

To evaluate the horizontal and vertical groundwater flow directions and the properties of the groundwater flow systems, I used the methods of basin hydraulics on a regional scale (TÓTH 2009). These methods are based on archive well data and involve compiling pressure-elevation profiles, tomographic potential maps, potential difference maps and a hydraulic cross-section.

Based on the results of the hydraulic evaluation, I outlined a conceptual groundwater flow model which was then further supported by the results of the hydrochemical analyses (major ion concentrations; ^{226}Ra, ^{222}Rn, total uranium activity; δ^2H and $\delta^{18}O$ stable isotope ratios) and 2D numerical simulation made by using the COMSOL Multiphysics software.

For laboratory analyses, water samples were collected from groundwater, surface water and a spring. During the sampling, the field parameters (temperature, pH, specific electrical conductivity, dissolved oxygen, redox-potential) were recorded on-site. The concentration of major ions (Ca^{2+}, Mg^{2+}, Na^+, K^+, HCO_3^-, Cl^-, SO_4^{2-}) and trace elements (e.g. U, Ba, Fe) were determined in the laboratories of the National Center for Public Health and Pharmacy and the Department of Geology (ELTE). Among the examined radionuclides, the activity concentrations of ^{226}Ra and total uranium (^{234}U+^{235}U+^{238}U) were measured in the Müller-Surbeck laboratory (Department of Geology, ELTE) by an innovative measurement technique used only at ELTE in Hungary, alpha spectrometer using selectively adsorbing Nucfilm discs. The ^{222}Rn activity concentration was measured at the Department of Atomic Physics (ELTE) using a TriCARB 1000TR liquid scintillation detector. The δ^2H and δ^{18}O measurements were performed in the isotope laboratory of the Finnish Geological Survey (GTK) in Espoo, using a PICARRO L2130 ^{2}H/^{18}O isotope analyser.

The investigation of the geochemical processes controlling the mobilization of uranium by groundwater was done through a 1D geochemical modelling task using the PHREEQC software.

3. Results, theses of the dissertation

Based on the combined evaluation of the results achieved by the methods used during my doctoral research, I came to the results and findings summarized in the following theses:

1) Based on the results of the regional-scale hydraulic evaluation carried out in the surrounding of the Velence Hills, I found that the driving force of the groundwater flow systems are the changes in the elevation of the groundwater table in the examined elevation range of (–250)–250 m asl.

- **a.** I found that above the 0 m asl elevation, the main horizontal water flow directions point from the Vértes, the Velence Hills and the loess ridges towards the Danube, Sárvíz Valley, Sárrét and Lake Velence. In the ((–250)–0 m asl elevation range, I observed only the discharge effect of the Sárvíz Valley and the Danube, while the effect of the local changes in topography disappeared.

b. Among the constructed 62 pressure-elevation profiles, 32 profiles indicated recharge areas characterised by downward groundwater flow. The recharge areas are characteristic of the elevated parts of the research area. The lowlands were identified as discharge regime areas. The potential difference maps and the hydraulic cross-section also proved that downward flow is the dominant vertical flow direction in the study area.

c. I observed that the profiles indicating the recharge and discharge areas alternate in a mosaic pattern, based on which I concluded the presence of local flow systems in the examined elevation interval.

2) Using the results of the hydrochemical analyses and the numerical modelling, I verified the conceptual model, which was outlined based on the results of the hydraulic evaluation.

a. Majority of the water samples collected from groundwater are characterized by low temperature, TDS and the dominance of HCO_3^-, while the δ2H and δ18O stable isotope ratios indicated an origin from precipitation in a recent climate. I identified these properties as evidence of local flow systems and recharge areas.

b. Based on the results of the 2D numerical groundwater flow simulation, I verified that the recharge and discharge areas alternate in a mosaic pattern along the cross-section. I observed a decrease in flow intensity with depth**,** which I explained by the decrease in hydraulic conductivity with depth.

3) I have revealed that there is regional geogenic uranium contamination in the groundwater in the study area.

a. I explained the presence of uranium by the groundwater flow conditions of the study area, i.e. by the existence of local flow systems characterized by shallow penetration depth, short residence time, oxidising environment and HCO_3^- dominance, and the dominance of recharge areas.

b. Based on the results of the activity concentration measurements of the most common radionuclides in drinking water (total uranium, ^{226}Ra and ^{222}Rn), I found that the gross alpha activity measured in drinking water exceeding the screening level of 0.1 Bq/l can be explained by the presence of uranium in the groundwater. The measured uranium activity values were below the derived concentration for uranium (^{234}U: 2.8 Bq/l and ^{238}U: 3 Bq/l). Therefore, no health risk arises from drinking water consumption.

4) Using geochemical modelling, I quantitatively and qualitatively verified the conceptual model explaining the spatial distribution of radionuclides, which was outlined based on the results of the hydraulic evaluation and hydrochemical analyses.

a. I identified the geochemical characteristics of the Pannonian-Quaternary siliciclastic aquifer system (redox potential, organic matter decomposition rate, carbonate content) that control the mobilization of uranium in groundwater.

b. I determined the possible date of the formation of the reducing environment (~560 years), which was approximately the same as the assumed residence time of groundwater.

c. I found that the PHREEQC software is suitable for modelling changes in the uranium content of groundwater and for quantifying the processes affecting the geochemical behaviour of uranium in rock-water systems.

5) I explored the relationship between Lake Velence and the groundwater flow systems: the lake is the discharge area of the groundwater flow systems.

a. I identified that groundwater flows towards Lake Velence from the Velence Hills, Gárdony and Székesfehérvár.

b. In the groundwater samples collected in the Bika-völgy in Gárdony, I measured total uranium activity of a similar order of magnitude to the values measured in the lake, which indicates the origin of the uranium content in the lake water.

c. Based on the similar hydrochemical characteristics of the groundwater and the lake, I confirmed that the typical geochemical character of soda lakes originates from the groundwater. This is also indicated by the dominant Mg^{2+} content in the groundwater, which is also characteristic of the lake's water.

d. Based on the results of the hydraulic evaluation and hydrochemical analyses, I found that the lake is fed by local flow systems, which results in the lake's vulnerability to anthropogenic and climate change impacts.

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