



Uranium in groundwater – The importance of hydraulic regime and groundwater flow system's understanding



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ABSTRACT

Uranium and other natural radionuclides are common components in groundwater, but they are not routinely measured. In drinking water their total activity is screened, but in the evaluation of the measured values usually the aquifer material is only considered. However, the occurrence of radionuclides in groundwater is strongly affected by flow systems and their geochemical characteristics. Therefore hydrogeology and flow system's evaluation is crucial to understand natural radioactivity. Areas of different hydraulic regimes – recharge, throughflow and discharge – even within the same aquifer are characterized by different geochemical environment. In the present study pressure-elevation profiles were generated based on existing basic hydraulic data of wells in order to determine the flow regimes and associated vertical groundwater flow directions. 24–753 mBq L⁻¹ uranium activity concentrations were found in groundwater and surface water showing great areal variability. High uranium values correspond to recharge regimes with downward flow directions. Uranium mobility is enhanced by high bicarbonate content and circumneutral pH. The study emphasize the importance of groundwater flow system's understanding in those areas, where elevated background radioactivity may exist.

1. Introduction

Moving groundwater is a geologic agent as it interacts with its environment (Tóth, 1999). It is able to mobilise, transport and accumulate matter, amongst others also radioactive elements. Therefore the members of the uranium and thorium decay series are common components in groundwater (Hoehn, 1998; Porcelli and Swarzenski, 2003; Skeppström and Olofsson, 2007). In case of the radioactivity assessment of groundwater, however, usually only the aquifer material or anthropogenic sources are considered (e.g. Birke et al., 2010; Bonotto, 2017; Liesch et al., 2015; Riedel and Kübeck, 2018) and the dynamics of groundwater and its role is often just neglected. However understanding the dynamic world of groundwater, organized into flow systems, is a key issue here, since the geochemical conditions are changing along the flow paths. Areas of different hydraulic regimes – recharge, throughflow and discharge – even within the same aquifer are characterized by different geochemical environment, from oxidizing to reducing, respectively. It is an important question e.g. in case of redox-sensitive parameters such as the uranium (Osmond and Cowart, 1999).

The hydrogeochemical behaviour of uranium is complex and depends to a great extent on a range of water quality parameters such as alkalinity, pH, redox conditions and chemical composition (Porcelli and Swarzenski, 2003; Siegel and Bryan, 2014). In groundwater the predominant species are in either U(IV) or U(VI) oxidation states (Sheppard, 1980; Ivanovich and Harmon, 1992; Chabaux et al., 2003). Under reducing conditions U(IV) is very insoluble and tends to precipitate (Gascoyne, 1992). In oxidizing environment uranium is in U(VI) state and as (UO₂²⁺) ion highly mobile (Sheppard, 1980; Chabaux, 2003). Its mobility can be enhanced by complexation by organic matter (e.g. humic and fulvic acids), carbonates, phosphates and hydroxides (Bourdon et al., 2003). Under circumneutral pH conditions complexes with carbonate and phosphate are dominant (Porcelli and Swarzenski, 2003), therefore uranium can be highly mobile especially in water where the pH is near neutral and has high carbonate alkalinity (Curtis et al., 2006). However, at circumneutral pH and in the absence of strong complexing agents, U(VI) transport is limited by strong adsorption on mineral surfaces in soils and sediments (Davis et al., 2004), while adsorption of uranium to colloids causes rather increased

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mobility (Porcelli and Swarzenski, 2003).

The research of natural radioactivity of groundwater is an important issue in those countries like Hungary, where both the drinking water supply and the bottled mineral water market rely almost 100% on groundwater (Somlai et al., 2002; Szerbin and Köteles, 2002; Kovács et al., 2004; Jobbágy et al., 2010). Following the Euratom Drinking Water Directive the radioactivity of drinking water is screened in Hungary by total alpha and total beta activity measurements. Such measurement initiated this study, where in case of one local waterwork the total alpha activity exceeded the 0.1 Bq L^{-1} threshold. Due to confidentiality agreement exact places and initial measured values cannot be shown. The regional Upper Pannonian sandy aquifer serves as water supply for many settlements in the region, where that elevated total alpha activity was reported, therefore a hydrogeological research was initiated to investigate the groundwater flow conditions in the region and evaluate the radionuclide content of groundwater in this context. Since the nuclide specific measurements pointed to the uranium as a main cause, in this paper we are focusing only on uranium. However, speciation of uranium isotopes are beyond the scope of this study, since we are interested in only what is dissolved in the water. The alpha spectrometry measurement method used gives the sum of ($^{234}\text{U} + ^{238}\text{U} + ^{235}\text{U}$) activities, with the ^{235}U activity making only about 5% of the ^{238}U activity. We will thus use the term ($^{238}\text{U} + ^{234}\text{U}$) for the above mentioned sum of the uranium isotopes. Activities given also always mean the above mentioned sum of the uranium isotopes. The term “uranium concentration” means the chemical uranium concentration determined by ICP-MS. It only gives the concentration of the ^{238}U . With a 4 orders of magnitude lower half-life the ^{234}U cannot be detected by standard ICP-MS.

Following the previous considerations above, as uranium mobile in oxidizing environments, recharge areas with downward flow conditions are those specific areas where uranium activity concentrations could be detected in groundwater if there is a source. In the study area the Upper Pannonian sandy aquifer and its covering sediments may contain fragments of granitic rocks of the nearby hills which are characterized by higher U content (2.5–5.4 ppm Burján et al., 2002), since in the past 2–3 million years the massive granite has been exposed to subaerial processes. Moreover, this uplifted hilly range may serve as a recharge area for groundwater in the region, and uranium could be leached out from the weathered rocks and transported by groundwater as well.

To determine the regime characteristics of an area i.e. recharge, throughflow or discharge, and the direction of groundwater flow, i.e. downward, lateral or upward, respectively, pressure-elevation profiles were generated based on archival well documentation available in public institutions. Additionally water samples were collected for chemical analysis, especially for uranium measurements, to evaluate the correlation between the type of regime and uranium activity concentrations and other physicochemical parameters.

2. Methods

2.1. Hydraulic data evaluation

Archival measured hydraulic data of wells were analysed by the basic regional hydraulic methods of hydrogeology (Tóth, 2009). Measured hydraulic data were taken from the archived original well documentation of government institutions. Essential basic data were (1) well coordinates, (2) elevation, (3) depth of the static water level, (4) elevation of the screened interval or measuring point. During the hydraulic calculations, the measured depth of water level was recalculated to hydraulic head by using the elevation. Afterwards hydraulic head data were converted to pore-pressure values with the help of the following equation, which assumes constant fluid density along the flow path:

$$h = z + p/(\rho \cdot g) \quad (1)$$

where h is the measured hydraulic head; z is the elevation of the measuring point with respect to the Baltic Sea level; p is pore-pressure; $(\rho \cdot g)$ is the specific weight of the fluid, which is numerically equal to the hydrostatic vertical pressure gradient of that fluid.

Based on the calculated pressures, pressure vs. elevation [$p(z)$] profiles were constructed, which *sensu stricto* allows for the examination of the vertical component of fluid flow directions by comparing the vertical pressure gradient to the ideal hydrostatic condition. The vertical pressure gradient (γ) is the change in pore pressure per unit vertical length in a flow domain. Ideally, in the hydrostatic condition there is no vertical fluid flow, because the buoyancy and the force of gravity on water particles are in balance. In this case the vertical pressure gradient is hydrostatic, and its average value is $\gamma_{st} = 0.981 \text{ MPa}/100 \text{ m}$, by applying the water density of 1000 kg m^{-3} . In the hydrodynamic state, a driving force can shift the system off balance, therefore the vertical flow component occurs too, and the vertical pressure gradient changes to dynamic (γ_{dyn}). In recharge areas $\gamma_{dyn} < \gamma_{st}$, so there is a downward flow direction and γ_{dyn} is called subhydrostatic, while in discharge areas $\gamma_{dyn} > \gamma_{st}$, thus there is an upward flow direction and γ_{dyn} is called superhydrostatic. In midline areas, the vertical pressure gradient is hydrostatic ($\gamma_{dyn} = \gamma_{st}$), therefore the fluid flow has no vertical component, nevertheless the horizontal component cannot be excluded.

On the $p(z)$ profiles beyond the data points, the (reference) hydrostatic vertical pressure gradient line (γ_{st}), the dynamic vertical pressure gradients (γ_{dyn}) fitted on the data points are also presented. An important prerequisite, that wells interpreted in one $p(z)$ profile had to have approximately the same land surface elevation, because position of the reference hydrostatic pressure gradient line depends on it. The area of the pressure-elevation profiles is also influenced by the areal and depth distribution of available hydraulic data.

2.2. Sampling and analytical methods

In-situ physicochemical parameters (specific electrical conductivity, temperature, pH, ORP, dissolved O_2) were recorded during sampling on the field by YSI Pro Plus multiparameter water quality instrument. Table 1 contains the accuracy of the device for each parameters.

Water samples were collected in 0.25 L polypropylene bottles for uranium measurements. For elemental analysis water samples were taken in 1.5 L PET bottles, 50 mL centrifuge tube and 0.25 L glass bottles. The samples were kept cool until delivery to the laboratories. The elemental analysis was performed in the laboratory of National Public Health Institute (Table 2). Uranium was measured both by ICP-MS (after nitric acid preparation) and with alpha spectrometry using Nucfilm discs, which are coated by selectively adsorbing thin films (Surbeck, 2000). The U-discs are made of polycarbonate, which is covered with epoxy resin fixing a finely ground ion exchange resin (Diphonix). For uranium measurements by alpha spectrometry the water samples were acidified with concentrated (85%) formic acid to pH 2 that guarantees that uranyl- CO_2 complexes are broken up. The U-discs were exposed to 100 mL acidified, stirred water for 20 h. After 20 h 90% of the uranium activity present in the sample is adsorbed. The

Table 1

Parameters, ranges and accuracy of YSI Pro Plus multiparameter water quality instrument.

Parameter	Accuracy	Range
T (ref. 25 °C)	0.2 °C	– 5 – 70 °C
DO	%: + - 2%, mg L^{-1} : + - 2%, temp. comp. -5 °C – 50 °C	0–500% 0–50 mg L^{-1}
EC	+ - 0.5%, 0.001 mS cm^{-1}	0–200 mS cm^{-1}
pH	0.2	0–14
ORP	+ - 20 mV	– 1999 – + 1999 mV

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