Science of the Total Environment 601–602 (2017) 679–690

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Temporal changes in radiological and chemical composition of Cambrian-Vendian groundwater in conditions of intensive water consumption

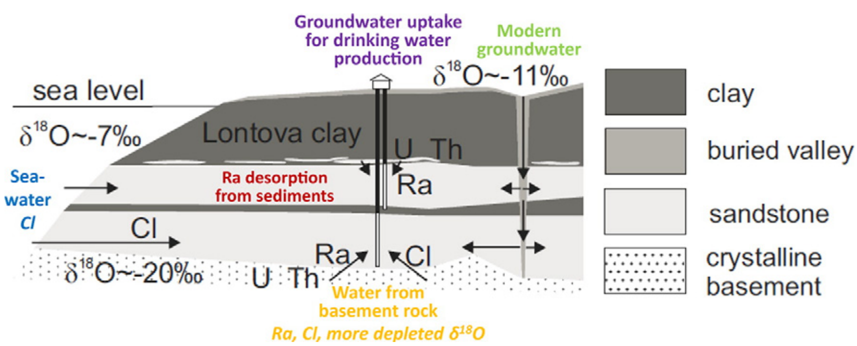
Siiri Suursoo [a,*], Liie Hill [b], Valle Raidla [c,d], Madis Kiisk [a], Alar Jantsikene [a], Nele Nilb [e], György Czuppon [f], Kaisa Putk [a], Rein Munter [b], Rein Koch [a], Kadri Isakar [a]

[a] University of Tartu, Institute of Physics, W. Ostwaldi Str. 1, 50411 Tartu, Estonia
[b] Tallinn University of Technology, Department of Chemical Engineering, Ehitajate tee 5, 19086 Tallinn, Estonia
[c] Tallinn University of Technology, Institute of Geology, Ehitajate tee 5, 19086 Tallinn, Estonia
[d] University of Heidelberg, Institute of Environmental Physics, Neuenheimer Feld 229, D-69120 Heidelberg, Germany
[e] Viimsi Vesi Ltd., Nelgi Str. 1, Viimsi parish, 74001 Harju County, Estonia
[f] Hungarian Academy of Sciences, Institute for Geological and Geochemical Research, Budaörsi út 45., H-1112 Budapest, Hungary.

HIGHLIGHTS

- Water uptake on a peninsula from Cambrian-Vendian aquifer was observed for 4 years
- Relevant radionuclides and chemical parameters (Cl, Mn, Fe, $\delta^{18}O$) were measured
- Inflow of meteoric water, water from the sea or from the basement rock may occur
- Cl and Ra monitoring are critical parameters for sustainable groundwater management
- Changes in Cl concentrations can be used to predict Ra activity concentrations

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 December 2016
Received in revised form 15 May 2017
Accepted 15 May 2017
Available online 31 May 2017

Editor: D. Barcelo

Keywords:
Groundwater management
Radionuclides
Fe
Mn
Cl
Stable isotope ratio ($\delta^{18}O$)

ABSTRACT

Intensive groundwater uptake is a process at the intersection of the anthroposphere, hydrosphere, and lithosphere. In this study, groundwater uptake on a peninsula where only one aquifer system – the Cambrian-Vendian (Cm—V) – is available for drinking water uptake is observed for a period of four years for relevant radionuclides and chemical parameters (Cl, Mn, Fe, $\delta^{18}O$). Intensive groundwater uptake from the Cm—V aquifer system may lead to water inflow either from the sea, through ancient buried valleys or from the under-laying crystalline basement rock which is rich in natural radionuclides. Changes in the geochemical conditions in the aquifer may in turn bring about desorption of Ra from sediment surface. Knowing the hydrogeological background of the wells helps to predict possible changes in water quality which in turn are important for sustainable groundwater management and optimization of water treatment processes. Changes in Cl and Ra concentrations are critical parameters to monitor for sustainable management of the Cm—V groundwater.
Radionuclide activity concentrations in groundwater are often considered rather stable, minimum monitoring frequency of the total indicative dose from drinking water is set at once every ten years. The present study demonstrates that this is not sufficient for ensuring stable drinking water quality in case of aquifer systems as

* Corresponding author.
E-mail address: siiri.suursoo@ut.ee (S. Suursoo).

http://dx.doi.org/10.1016/j.scitotenv.2017.05.136

sensitive as the Cm—V aquifer system. Changes in Cl concentrations can be used as a tool to predict Ra activity concentrations and distribute the production between different wells opening to the same aquifer system.

1. Introduction

The Cambrian-Vendian (Cm—V) aquifer system is the lowest aquifer system available for drinking water uptake in Estonian territory. It lies on the crystalline basement rock known to be rich in uranium and thorium (Raudsep, 1997). The aquifer system is an important drinking water reservoir in northern Estonia (Karise, 1997). Intensive groundwater uptake may lead to water inflow either from the sea (a), from the underlying crystalline basement rock (b) or through ancient buried valleys (c) (Marandi et al., 2012). This in turn may bring about many changes in water quality, including the alteration of radiological parameters. Significant changes may lead to the need to make adjustments in the water treatment process to be able to meet the criteria for drinking water quality.

The groundwater from the Cm—V aquifer system on the Viimsi peninsula contains up to 600 mBq/L of ^{226}Ra and 800 mBq/L of ^{228}Ra. Such ^{226}Ra and ^{228}Ra activity concentrations in drinking water lead to an effective dose above 0.5 mSv/yr, which is 5 times higher than the guidance level suggested by the International Commission on Radiological Protection (ICRP, 2000) and the World Health Organization (WHO, 2011). In principle, the intake of radionuclides from drinking water consumption can be classified as a planned exposure situation or as an existing exposure situation for the public. As a conservative approach, ICRP suggests to use the dose constraint of the former – 0.1 mSv/yr – as the guidance level for effective dose from drinking water consumption (ICRP, 2000). Exceeding the value should not be taken as an indication that the drinking water is unsafe, but as a trigger for further investigation (WHO, 2011).

0.10 mSv/yr has been adopted by the Directive 2013/51/EURATOM as the parametric value for the total indicative dose (TID) from drinking water consumption. In case of non-compliance a European Union member state should consider whether it poses a risk to human health which requires action and, where necessary, take remedial action to improve the quality of the water to a level which complies with the requirements for the protection of human health from a radiation protection point of view (Council of the European Union, 2013).

Radium is not chemically inert in water. Its presence does not depend only on the existence of the parent nuclides but also on dissolving and precipitation processes governed by the chemical properties of the environment (Chau et al., 2011; IAEA, 2014). Therefore, changes in the water source may cause fluctuations of radium activity concentrations which are not dependent on the concentration of parent nuclides.

The problem of high radium activity concentrations in the Cm—V aquifer system has been known for more than two decades (Estonian Radiation Protection Centre, 2005; Kiisk et al., 2011). Today, quite a lot of data are available on radium content of groundwater from the Cm—V aquifer system (Savitskaja and Viigand, 1994; Savitskaja and Jaštšuk, 2001; Forte et al., 2010). However, there is still limited knowledge of the other radionuclides in Estonian groundwater and hardly any information has been published on changes in radium activity concentrations in the same well over a longer period of time.

In this study, nine Cm—V wells located on the Viimsi peninsula, Estonia (Fig. 1) were observed from 2012 to 2015. The wells were drilled in 2008–2009, extensive groundwater uptake started in 2012 with a commissioning of a new water treatment facility planned for serving *ca* 15,000 people with drinking water. Maximum designed output capacity of the treatment plant is 6000 m^3/day. In reality, the average output capacity has stayed around 2600 m^3/day.

The objective of this work was to monitor relevant radionuclides (radium isotopes), chemical parameters (Cl, Mn, Fe), and stable isotope ratios of oxygen and hydrogen (δ^{18}O, δD) in groundwater in order to detect changes in well water quality in a situation of intensive uptake. Trends in Ra, Cl, Mn, Fe, δ^{18}O, and δD values, in combination with information on regional hydrogeology, were used to make inferences about the causes of observed water quality changes. This is a critical input to be able to manage the groundwater resource sustainably. Suggestions are given on what to consider when developing a radionuclide monitoring strategy for the wells. The research should help the water treatment facility operator to plan changes in the treatment technology to be able to meet the criteria for drinking water. More generally, the hydrogeological effects taking place on the Viimsi peninsula should be considered as a possible scenario in other locations with a similar hydrological situation – wells which are sensitive to water uptake. The present work demonstrates that in sensitive systems general monitoring strategies may not be adequate. Instead, the local hydrogeological situation should be taken into account when setting the maximum allowed production rates for wells and developing a monitoring plan for water quality.

2. The study area

The Viimsi peninsula is located in the north-east of Tallinn, capital of Estonia (Fig. 1). The length of the peninsula is 15 km and the average width about 5 km. The landscape is flat and its surface consists of glacial and marine deposits from the Quaternary Period. The peninsula is underlain by Ordovician carbonate and Cambrian clastic sedimentary rocks. The majority of the local population lives on the coast. During the last decade, that population has tripled due to active real estate development and it has now reached 19,000 inhabitants. This has caused problems with potable water supply.

The fact that the Cm—V aquifer system represents the only significant source of drinking water on the peninsula makes the situation unique. For the time being there are also no possibilities to permanently import consumer water from inland. In principle, groundwater from the Ordovician-Cambrian aquifer system and Quaternary sediments can be used, but their amount is rather limited and their role in the public water supply system is insignificant.

The Cm—V aquifer system is well confined from overlying aquifers, intrusion of meteoric water, and surface pollution by the Lontova-Lükati aquitard. However, in some places in North Estonia, the aquitard is cut by deep pre-Quaternary valleys filled with sediments with higher hydraulic conductivity. In these regions, the Cm—V aquifer system is exposed to upper groundwater aquifers (Raidla et al., 2009). The groundwater from the Cm—V aquifer system in North Estonia has typically more depleted ^{18}O values (−18 to −23‰; Raidla et al., 2009) than local modern precipitations (−10.4‰; Punning et al., 1987) and shallow groundwaters (−11 to −12.5‰; Raidla et al., 2016). This phenomenon could be explained by the fact that the Cm—V groundwater originates from glacial meltwater, since Scandinavian ice sheet covered the territory of Estonia during the last ice age (Vaikmäe et al., 2001; Raidla et al., 2012). Variations in the geochemical and stable isotope composition suggest that the current water in the Cm—V aquifer system is a mixture of three components – glacial melt water, brine which was present in the aquifer system before the intrusion of the glacial water, and recent meteoric water which has intruded into the system through deep buried valleys (Raidla et al., 2009).