RADIOCARBON AND OTHER ENVIRONMENTAL ISOTOPES IN THE GROUNDWATER OF THE SITES FOR A PLANNED NEW NUCLEAR POWER PLANT IN LITHUANIA

Jonas Mažeika[1] • Tõnu Martma[2] • Rimantas Petrošius[1] • Vaidotė Jakimavičiūtė-Maselienė[1,3] • Žana Skuratovič[1,4]

ABSTRACT. The assessment of construction sites for the new Visaginas Nuclear Power Plant (Visaginas NPP), including groundwater characterization, took place over the last few years. For a better understanding of the groundwater system, studies on radiocarbon; tritium; stable isotopes of hydrogen, oxygen, and carbon; and helium content were carried out at the location of the new NPP, at the Western and Eastern sites, as well as in the near-surface repository (NSR) site. Two critical depth zones in the Quaternary aquifer system were characterized by different groundwater residence times and having slightly different stable isotope features and helium content. The first shallow interval of the Quaternary multi-aquifer system consists of an unconfined aquifer and semiconfined aquifer. The second depth interval of the system is related to the lower Quaternary confined aquifer. Groundwater residence time in the first flow system was mainly based on tritium data and ranges from 6 to 60 yr. These aquifers are the most important in terms of safety assessment and are considered as a potential radionuclide transfer pathway in safety assessment. Groundwater residence time in the lower Quaternary aquifers based on ^{14}C data varies from modern to several thousand years and in some intervals up to 10,500 yr.

INTRODUCTION

Lithuania's obligation to shut down units 1 and 2 of the Ignalina Nuclear Power Plant (INPP) and to decommission them as soon as possible was one of the requirements included in the European Union Accession Treaty. Unit 1 was shut down on 31 December 2004 and Unit 2 on 31 December 2009. The low- and intermediate-level radioactive waste (LILRW) produced during the operation and decommissioning of the INPP will be disposed in a near-surface repository (NSR) located near the INPP site. After the shutdown of the INPP, formerly the principal electricity producer in Lithuania, there are plans to build the new Visaginas NPP located near the existing INPP site.

The new NPP sites and the NSR site are located in the northeastern part of Lithuania on the shore of Lake Druksiai, close to the borders of Belarus and Latvia (Figure 1). One site (Eastern) is located directly east of the existing INPP and the other (Western) is located west of the INPP. The NSR site is located about 1 km south of the INPP.

In the last few years, investigations related to the assessment of construction sites for the new NPP were rather intensive. To better understand the groundwater system, preliminary studies on radiocarbon (^{14}C), tritium (^{3}H), stable isotopes of hydrogen, oxygen, and carbon (^{2}H, ^{18}O, ^{12}C), and helium (^{3}He+^{4}He) content were carried out at all the studied sites. The regional geological and hydrogeological settings allow us to distinguish 2 regional aquifer systems in the groundwater of the active circulation zone of the whole region: the Quaternary aquifer system and the Upper-Middle Devonian aquifer system (Juodkazis 1979).

The Quaternary aquifer system in the eastern part of Lithuania occupies an area of ~20,880 km^2, i.e. approximately one third the territory of Lithuania. In a regional context, the Quaternary aquifer system is the first water-bearing hydrogeological system from the top in the INPP region. This system

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[1] Nature Research Centre, Vilnius, Lithuania, Akademijos str. 2, LT–08412 Vilnius, Lithuania.
[2] Tallinn University of Technology, Institute of Geology, Ehitajate tee 5, 19086 Tallinn, Estonia.
[3] Vilnius University, M.K. Čiurlionio str. 21/27, LT-03101 Vilnius, Lithuania.
[4] Corresponding author. Email: skuratovic@geo.lt.

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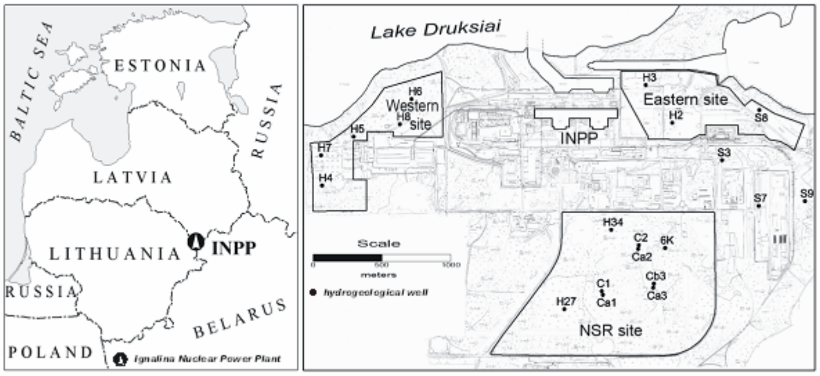

Figure 1 Location of the study area

contains an unconfined aquifer and a series of semiconfined and confined aquifers attributed to intermoraine deposits of sandy and gravel material from different interglacial periods. In the present study, the uppermost Quaternary aquifer is treated as semiconfined mainly due to the very small height of hydraulic head above the top boundary of the aquifer. The remaining several Quaternary aquifers are confined with the hydraulic head clearly above the top boundary of aquifers. The average thickness of the Quaternary succession is 85–105 m and reaches up to 260 m in paleovalleys (Marcinkevičius and Laškovas 2007).

The Middle-Upper Devonian multi-aquifer system (>50,000 km^2) in Lithuania is a very important groundwater body for the municipal and domestic supply of fresh and mineral water (Mokrik et al. 2008). The aquifer system is a classical confined system with a high hydraulic head above the top boundary and is composed of many sublayers of different hydraulic conductivity.

The present study is focused on the uppermost part of the Quaternary aquifer system including unconfined groundwater, semiconfined aquifer (fIIInm3), and confined aquifers (fIImd, fIIžm-dn) that occur at all 3 sites (Juodkazis 1979). The unconfined groundwater and semiconfined aquifers are separated by a layer of clay material. They are most important for safety assessment and could act as a radionuclide pathway considering different scenarios of the site's evolution.

METHODS

The groundwater for analysis of ^{14}C-specific activity in dissolved inorganic carbon (DIC) was sampled into 80-L vessels in volumes sufficient for analysis (240–320 L). All DIC components were precipitated using appropriate quantities of NaOH and $CaCl_2$ (Arslanov 1985). After drying the precipitated carbonate material in the laboratory, this material was separated into appropriate proportions for the ^{14}C and δ^{13}C analysis. A conventional method was applied for benzene synthesis from carbonates (Arslanov 1985; Gupta and Polach 1985). The ^{14}C-specific activity of benzene was measured by a liquid scintillation analyzer (Tri-Carb® 3170TR/SL). The main performance parameters of the spectrometric system for ^{14}C measurement in benzene form (~3 g) with 7-mL low-background glass vials were better than the background count rate (0.65 ± 0.05 CPM) and counting efficiency (71.4 ± 0.8%). The results are presented as specific activity (percent of Modern Carbon: 1 pMC = 2.27 Bq/kg C) (Stuiver and Polach 1977). The radiometric accuracy of measurements was better than 1%. The quality of ^{14}C determinations was periodically tested through participating in various intercomparison studies. The results of these intercomparisons (FIRI and VIRI programs;

Scott et al. 2003, 2010), indicated that most of the ^{14}C results within 1σ are of 3% precision, with some reaching 1.5% precision.

Groundwater samples for tritium (^{3}H) measurements were collected in 0.6-L plastic bottles. The beta activity counting of ^{3}H was run for a scintillation cocktail with 12 mL of Ultima Gold™ LLT and 8 mL of water electrolytically enriched with ^{3}H. Measurements were performed with the Quantulus 1220™ liquid scintillation spectrometer. The main performance parameters of the spectrometric system for ^{3}H with 20-mL plastic vials were as follows: background count rate was 1.0 ± 0.1 CPM and counting efficiency was 22.0 ± 0.4%. The detection limit for ^{3}H was 0.1 TU (1 TU corresponds to 0.118 Bq/L). The normal precision of ^{3}H measurements in the water was recognized in the comparison, organized by the Latvian Environment Agency Laboratory, in the Seventh IAEA intercomparison of low-level tritium measurements in water (TRIC2004) and in the Nordic Nuclear Safety Research Intercomparison of Laboratory Analyses of Radionuclides in Environmental Samples (NKS LABINCO) Intercomparison Exercise 2004–2005.

The stable carbon isotope ratio ($^{13}C/^{12}C$) of groundwater DIC was expressed in $\delta^{13}C$ values relative to the international VPDB standard (Coplen et al. 2006). Analysis using the gas source isotope ratio mass spectrometry (IRMS) method was performed by analyzing extracted CO_2 from groundwater carbonates using a Thermo Fisher Scientific Delta V Advantage and GasBench II system. $^{18}O/^{16}O$ and $^{2}H/^{1}H$ ratios of water molecules were determined as $\delta^{18}O$ and $\delta^{2}H$ values with respect to the international standard (VSMOW) (Coplen 1996), using a IRMS Delta V Advantage and GasBench II system or Picarro L2120-i Cavity Ringtown Spectrometer. Stable isotope data are presented as per mil deviations from internationally accepted standards with the reproducibility of ±0.1‰ for ^{18}O, ^{13}C and ±1‰ for ^{2}H. The concentration of helium (the sum of isotopes ^{3}He and ^{4}He) present in the groundwater (percent of helium in gases dissolved in water) was measured by the helium indicator INGEM-1 following the quartz membrane and magnetic discharge principle (Yanitsky 1979). The radioactive isotopes and helium measurements were performed in the Radioisotope Research Laboratory of Nature Research Centre, Vilnius, Lithuania, and stable isotope measurements in the Laboratory of Isotopes Palaeoclimatology of the Institute of Geology of Tallinn University of Technology, Estonia.

RESULTS

The basic approach of the study was to use the radioisotope and stable isotope data (Table 1) as well as the groundwater chemistry data (Table 2) to define groundwater residence time and other hydrodynamic features of top aquifers attributed to the Quaternary aquifer system of the studied sites. The groundwater chemistry data for this study were kindly provided by the Joint Stock Company (JSC) Visaginas NPP.

The ^{3}H activity in the groundwater of the studied area can be related to ^{3}H variation in atmospheric precipitation and in surface water as groundwater recharge sources. The mean annual ^{3}H activity in atmospheric precipitation in the last 10 yr is about 10 TU (Mažeika et al. 2009). Significantly higher ^{3}H activity (an average of 40–50 TU and maximum of 204 ±11 TU in 2003) can be observed in the water of Lake Druksiai and boggy areas on the lake shores that are related with pathways of liquid releases from the INPP in normal operation. After the shutdown of INPP, the ^{3}H activity in the lake water has been continuously reduced to 12.7 ± 0.8 TU in 2012. The ^{3}H activity in the groundwater from unconfined and semiconfined aquifers is close to that of atmospheric precipitation and in some cases exceeds atmospheric values (Table 1). The maximum value of ^{3}H activity in the groundwater was determined in the NSR site borehole Ca-2 (screening depth 10.0–19.0 m) and was 22.2 ± 4 TU. The ^{3}H activity in the groundwater from the confined aquifer mostly is lower than in atmospheric precipitation and in some cases below the detection limit (<0.1 TU). The groundwater with no ^{3}H occurs at depths greater than 25–30 m.

The bicarbonate ion is the dominant form of DIC and ^{14}C in the groundwater. The ^{14}C of DIC is susceptible to reaction and dilution with dead carbon from carbonates and organic matter. The ^{14}C concentrations in the groundwater show a wide range of values from 15.5 pMC (Western site; confined aquifer fIIžm-dn; screening depth 61–65 m) to 94.3 pMC (Eastern site; confined aquifer fIImd; screening depth 9.6–13.2 m) in a rather small area (Table 1).

Helium concentration in the groundwater is mostly of atmospheric level (5.2×10^{-5} mL/L at STP, standard temperature and pressure). Only in some boreholes located in the Western site (H5, screening depth 84–88 m; H4, screening depth 129–134 m), helium content is slightly above the atmospheric level and reaches 16.0×10^{-5} mL/L STP, suggesting He inflow from the deeper aquifer to the Quaternary aquifer system in the Western site. However, there is no reason to relate these He traces in the groundwater with the occurrence of the hydrogeologically active tectonic zone. He content distribution in the 3 studied sites is rather uniform and not anomalous (Table 1).

The global meteoric water line (GMWL), satisfying the equation,

$$\delta^{2}H = 8\delta^{18}O + 10 \tag{1}$$

describes the relation between hydrogen and oxygen isotope composition of worldwide precipitation (Craig 1961). The average value of modern deuterium excess (d excess) for precipitation is 10. The d excess, $d = \delta^{2}H - 8 \times \delta^{18}O$ (Dansgaard 1964), reflects the kinetic fractionation effect and is a useful tool for identifying the origin of precipitating atmospheric moisture. A preliminary relationship between $\delta^{2}H$ and $\delta^{18}O$ for precipitation (monthly samples of 2010 and 2011 from 2 sampling sites) in east Lithuania was determined in terms of the local meteoric water line (LMWL) (Figure 2):

$$\delta^{2}H = 7.8\delta^{18}O + 7.2 \tag{2}$$

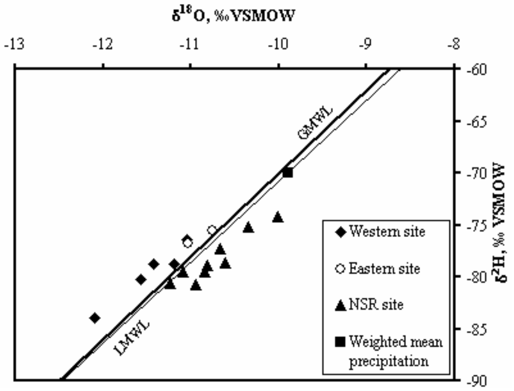

Figure 2 Stable isotope composition of investigated groundwaters.

The weighted mean annual $\delta^{18}O$ and $\delta^{2}H$ values for atmospheric precipitation are –9.9‰ and –70‰, varying respectively from –12.1 to –9.3‰ and from –82 to –70‰. The majority of groundwater samples in the ^{18}O/deuterium diagram are situated near the GMWL and LMWL, indicating groundwater recharge by modern atmospheric precipitation. The most positive $\delta^{18}O$ values are slightly below the GMWL and LMWL and are characteristic of shallow groundwater formed by modern

Table 1 Radioisotopes and stable isotopes data. Groundwater residence time in aquifer based on ^{3}H data (GRT (^{3}H)) is calculated for piston flow conditions and for α = 0.66. Groundwater age with δ^{13}C correction is derived from Equation 3. Groundwater age with correction based on ^{3}H data is done according to Equation 4.

Site	Well nr	Screening depth (m)	He content, 10^{-5} mL/L at STP	δ^{18}O (‰)	δ^2H (‰)	^{3}H, TU (±1σ)	GRT (^{3}H), yr	pMC (±1σ)	^{14}C age uncorrected, yr	δ^{13}C (‰)	^{14}C age (δ^{13}C), yr	^{14}C age (^{3}H), yr
Western												
	Semiconfined aquifer (fIIInm3)											
	H6	22.0–27.0	5.2	–11.4	–78.8	8.5 ± 0.3	32	74.2 ± 0.5	2500 ± 50	–13.0	Modern	Modern
	H8	22.0–25.0	5.2	–11.6	–80.2	9.0 ± 0.3	32	77.9 ± 0.5	2100 ± 50	–17.4	Modern	Modern
	Confined (fIIžm-dn)											
	H7	61.0–65.0	5.2	–12.1	–84.0	<0.1	>58	15.5 ± 0.3	15,400 ± 160	–7.4	8300	10,500
	H5	84.0–88.0	16.0	–11.2	–78.8	<0.1	>58	25.2 ± 0.3	11,400 ± 90	–13.7	9300	6500
	H4	129.0–134.0	10.0	–11.1	–76.4	<0.1	>58	32.0 ± 0.4	9400 ± 90	–15.1	8200	4500
Eastern												
	Confined aquifer (fIImd)											
	H3	9.6–13.2	5.2	–10.8	–75.6	12.3 ± 0.4	35	94.3 ± 0.5	500 ± 50	–16.8	Modern	Modern
	H2	21.6–23.6	5.2	–11.0	–76.8	1.8 ± 0.1	57	62.2 ± 0.6	3900 ± 70	–14.1	Modern	Modern
	Unconfined groundwater											
NSR												
	6k	12.5–15.0	5.2	–11.5	n/d	9.9 ± 0.3	33	74.7 ± 0.7	2410 ± 80	–14.1	600 ± 200	Modern
	Semiconfined aquifer (fIIInm3)											
	Ca-1	9.0–22.0	5.2	–11.2	–80.6	13.4 ± 3.8	36	73.7 ± 0.9	2500 ± 100	–15.7	1600 ± 225	Modern
	Ca-2	10.0–19.0	5.2	–11.1	–79.5	22.2 ± 4.0	44	91.0 ± 1.0	780 ± 90	n/d	n/d	Modern
	Ca-3	11.0–21.0	5.2	–10.8	–79.5	11.9 ± 0.4	35	88.2 ± 1.2	1035 ± 110	–16.0	270 ± 220	Modern
	S7	12.2–15.2	5.2	–10.0	–74.2	18.9 ± 3.9	43	77.7 ± 0.9	2090 ± 90	–15.0	790 ± 200	Modern
	S9	12.2–15.2	5.2	n/d	n/d	5.4 ± 0.3	6	70.0 ± 0.9	2950 ± 110	n/d	n/d	Modern
	S3	12.7–15.7	5.2	–10.7	–77.3	7.9 ± 3.5	32	60.4 ± 0.9	4170 ± 120	–13.8	2180 ± 240	Modern
	Confined aquifer (fIImd)											
	S8	17.2–20.6	5.2	–10.3	–75.2	1.2 ± 0.2	57	54.3 ± 0.7	5045 ± 110	–13.9	3120 ± 220	Modern
	C-2	23.5–28.0	5.2	–11.1	–79.5	5.3 ± 3.4	6	65.6 ± 1.0	3500 ± 120	–14.0	1600 ± 240	Modern
	Cb-3	23.9–28.4	5.2	–11.0	–80.7	3.5 ± 0.2	26	56.5 ± 0.7	4700 ± 110	–14.5	3150 ± 220	Modern
	C-1	23.6–30.6	5.2	–10.8	–78.9	<0.1	>58	56.6 ± 0.7	4700 ± 100	–12.7	2030 ± 220	Modern
	H27	24.2–27.8	5.2	–11.1	n.d	<0.1	>58	56.4 ± 0.6	4740 ± 80	–8.4	Modern	Modern
	H34	33.6–36.0	5.2	–10.9	n.d	1.7 ± 0.1	57	37.5 ± 0.5	8100 ± 100	–7.0	520 ± 225	1900

Table 2 Hydrochemical data of studied groundwater.

Site	Well nr	Screening depth (m)	Cl^-	SO_4^{2-}	HCO_3^-	Na^+	K^+	Ca^{2+}	Mg^{2+}	TDS mg/L	Permanganate mg O/L	pH	ChOD mg O/L	CO_2 (aggressive) mg/L	CO_2 (equilibrium)
Western															
	Semiconfined aquifer (fIIInm3)														
	H6	22.0–27.0	4.2	14.7	371	11.6	1.5	87.7	22.4	514	3.9	7.4	10.0	<1.0	29.2
	H8	22.0–25.0	7.7	17.4	468	8.5	1.7	117.0	30.2	655	2.4	7.3	29.3	<1.0	45.1
	Confined (fIIžm-dn)														
	H7	61.0–65.0	2.1	<1.0	471	45.4	1.9	77.8	27.3	627	3.1	7.6	11.2	<1.0	23.8
	H5	84.0–88.0	3.7	<1.0	391	37.6	2.1	71.4	20.8	528	5.1	7.5	24.3	2.2	26.5
Eastern															
	Confined aquifer (fIImd)														
	H3	9.6–13.2	23.2	8.6	432	22.5	1.8	97.8	29.0	615	4.2	7.4	17.1	1.6	34.4
	H2	21.6–23.6	5.1	42.2	616	18.4	2.2	154.0	37.8	877	5.0	7.1	16.4	4.0	96.9
NRS															
	Unconfined groundwater														
	6k	12.5–15.0	77.5	51.4	788	31.0	5.5	207.0	62.5	1223	2.1	6.8	22.3	17.6	n/m[a]
	Semiconfined aquifer (fIIInm3)														
	Ca-1	9.0–22.0	19.9	20.4	463	9.9	1.4	124.0	34.3	681	1.3	7.4	4.1	n/m	16.2
	Ca-2	10.0–19.0	4.3	36.3	401	4.4	1.3	107.0	29.5	587	1.6	7.4	4.5	n/m	29.5
	Ca-3	11.0–21.0	10.0	48.2	502	6.6	1.8	136.0	35.5	743	1.5	7.6	6.7	n/m	22.9
	S7	12.2–15.2	3.6	<1.0	404	12.5	2.3	93.3	24.2	547	4.7	7.5	14.9	n/m	27.8
	S9	12.2–15.2	12.0	<1.0	447	11.9	2.3	106.0	27.5	613	3.9	7.2	11.6	n/m	57.2
	S3	12.7–15.7	6.6	3.3	340	7.0	1.7	81.8	21.8	467	3.2	7.3	8.8	n/m	31.9
	Confined aquifer (fIImd)														
	S8	17.2–20.6	2.5	<1.0	392	13.7	2.0	85.1	24.7	527	5.2	7.4	18.5	n/m	34.7
	C-2	23.5–28.0	2.7	<1.0	276	8.0	1.4	65.2	16.8	374	4.9	7.6	11.6	n/m	16.1
	Cb-3	23.9–28.4	4.4	6.4	304	9.7	1.6	61.6	17.6	409	2.6	7.6	9.3	n/m	16.2
	C-1	23.6–30.6	2.1	<1.0	318	11.2	1.5	71.8	18.9	428	5.0	7.6	11.6	n/m	17.2
	H27	24.2–27.8	1.0	1.6	387	13.7	2.1	104.0	25.0	535	23.2	7.6	n/m	n/m	21.1
	H34	33.6–36.0	8.7	1.6	356	23.6	2.2	82.3	22.3	497	5.0	8.1	n/m	n/m	6.3

[a] n/m = not measured.

precipitation and mixed with the water that is evaporated on the surface or during the infiltration through the unsaturated zone before reaching the aquifer. The most negative $\delta^{18}O$ values (reaching –12.1‰) refer to deeper confined groundwater and indicate the presence of water recharged in colder climatic conditions (glacial meltwater) (Boulton et al. 1999).

Based on hydrochemistry data (Table 2, Figure 3), the groundwater of the studied sites is of calcium-magnesium-bicarbonate type with total mineralization (TDS = total dissolved solids) equal to 467–745 mg/L in the semiconfined aquifer (fIIInm3) and 374–643 mg/L in the confined aquifer (fIImd). In the Piper trilinear diagram in Figure 3, the 2 lower triangles show the percentage distribution, in milliequivalents, of the major cations (Ca^{2+}, Mg^{2+}, Na^+, K^+) and major anions (SO_4^{2-}, Cl^-, CO_3^{2-} + HCO_3^-). The diamond shaped part of the diagram summarizes the dominant cations and anions. The final groundwater type is a mixed Ca^{2+}–Mg^{2+} type with highly prevailing weak acids HCO_3^- compared to strong acids (SO_4^{2-} and Cl^-).

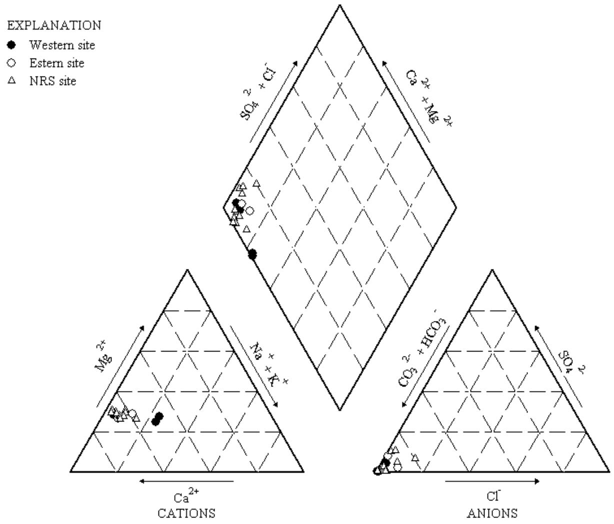

Figure 3 Piper diagram of studied groundwater samples

The chemical composition of groundwater from the deeper confined aquifer (boreholes H5, H7, H34) stands out of the general context and is a calcium-sodium-bicarbonate type with total dissolved solids (TDS) content varying from 497 to 627 mg/L. The chemical composition of groundwater from the shallow unconfined and semiconfined aquifers (boreholes 6k, Ca-1, H3, and H8) point to a hydraulic connection with the surface water.

The high content of aggressive CO_2 in the shallow unconfined groundwater (borehole 6k) indicates a hydraulic connection with the unsaturated zone and input of CO_2 from root respiration and decomposition of organic matter. More negative $\delta^{13}C$ values ($\delta^{13}C$ = –18.5‰ at NSR borehole S1, screening depth 12.7–15.7 m, and $\delta^{13}C$ = –17.4‰ at the Western site, borehole H8, screening depth 22–25 m) indicate an open system with respect to gaseous CO_2 in the groundwater. More positive $\delta^{13}C$ values are features of closed groundwater systems (Mokrik et al. 2008). This is characteristic of groundwater from the Western site (borehole H7, screening depth 61–65 m, $\delta^{13}C$ = –7.4‰) and from the NSR site (borehole H34, screening depth 33.6–36.0 m, $\delta^{13}C$ =–7.0‰) with deeper occurrences of groundwater.

DISCUSSION

Groundwater Age and Mean Residence Time

In this article, ^{3}H, ^{13}C and ^{14}C data are used to determine the groundwater age characteristics that depend on the initial ^{14}C activity. Geochemical models for the estimation of initial ^{14}C activity of groundwater are based on different assumptions about processes occurring in both unsaturated and saturation zones. They are well-known statistical models (Vogel 1970) based on alkalinity (Tamers 1975), chemical mass balance (Clark and Fritz 1997), carbon mixing (Pearson 1965; Pearson and Hanshaw 1970; Mook 1972; Wigley 1976), matrix exchange (Fontes and Garnier 1979). All these models consider the initial specific activity of ^{14}C and its modification by the geochemical processes estimated for a certain reservoir in term of the dilution factor (q).

For the investigated groundwaters, the dilution factor q was determined using δ^{13}C values of DIC (Ferronsky and Polyakov 1982, 2012) and ^{3}H data (Verhagen et al. 1991). The approach for dilution factor q determination (Ferronsky and Polyakov 1982, 2012) was used for groundwater dynamics characterization in the Baltic artesian basin (Mokrik et al. 2008) and is presented for comparison in this study as well. In Ferronsky and Polyakov (2012), the following formula for groundwater dating by ^{14}C is employed:

$$t_{age} = 8267 \ln\left(-\frac{5.7 \cdot \delta^{13}C_{meas}}{^{14}C_{meas}}\right) \tag{3}$$

where $^{14}C_{meas}$ and $\delta^{13}C_{meas}$ are the ^{14}C-specific activity and δ^{13}C values measured in DIC of groundwater.

This formula considers congruent dilution of carbonates and initial ^{14}C activity in recharge input taking into account 2 end-members (modern soil CO_2 and old dissolved carbonates) with ^{13}C and ^{14}C fractionation in the gas-fluid system. Applicability of this formula can be formally checked using a ^{14}C–^{13}C graph as proposed by Ferronsky and Polyakov (1982). Applying Equation 3, uncorrected ^{14}C ages are transformed to corrected ages that are significantly younger (Table 1).

The tritium-bearing groundwater age, or mean residence time of groundwater, in aquifers can be assessed based on ^{3}H data and a lumped parameter approach. The theory of the lumped parameter model was developed by Maloszewski and Zuber (1982) as a tool for evaluation of the mean residence time of groundwater. Details about the application of lumped parameter models for calculating the mean residence time of groundwater can be found in Maloszewski (1996). The lumped parameter models are represented by several computer codes, namely FLOWPC (Maloszewski 1996), BOXMODEL (Zoellmann et al. 2001), and TRACER (Bayari 2002). This study employs the BOXMODEL for ^{3}H data evaluation. The mean residence time of groundwaters was first estimated assuming a piston flow model as the calculations were based on only single tritium measurements.

The ^{3}H concentration in monthly precipitation records since 1952 is based on correlation of the Global Network of Isotopes in Precipitation (GNIP Vienna Hohe Warte station) data with the ^{3}H measurements in month precipitation in east Lithuania since 1999 (Mažeika et al. 2009). From the correlated monthly record of ^{3}H concentrations in precipitation, the ^{3}H input function was set to $\alpha = 0.66$, where $\alpha = \alpha_s/\alpha_w$ and represents the ratio of infiltration coefficients in summer (α_s) and winter (α_w) months of each year. The α value was derived from local precipitation and δ^{18}O data (however only for 2 yr) and δ^{18}O of shallow groundwater (Grabczak et al. 1984).

Using the measured ^{3}H data, the selected ^{3}H input function, and assuming a piston flow condition, mean groundwater residence times were calculated. As the ^{3}H input curve is not monotonic, several residence time values are feasible for a single ^{3}H measurement. For instance, with ^{3}H concentration 5.4 TU in groundwater, the following residence time values are possible: 6, 13, 29, 49, 55, and 56 yr. In order to follow a conservative approach, the youngest age values from several solutions are included in Table 1.

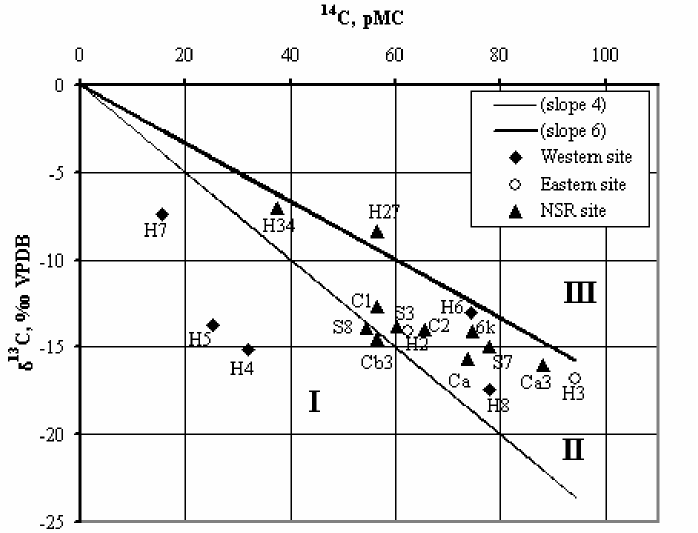

Figure 4 Relationship between ^{14}C content and δ^{13}C values of DIC in studied groundwaters. As derived in Ferronsky and Polyakov (2012), the solid line (slope 6) passes through the point with coordinates 102 pMC and $\delta^{13}C = -17‰$, being close to equilibrium state of carbon isotope exchange in gas-fluid system at fractionation factors for δ^{13}C and ^{14}C equal to 1.0070 and 1.0016 and isotope features of modern soil CO_2 with $\delta^{13}C = -25‰$ and $A^{14}C = 100$ pMC (slope 4). The experimental points located in zone II or close to it would correspond to modern or close-to-modern groundwater, the ^{14}C-specific activity decrease in DIC of which is due to dilution with the old dissolved carbonates ($\delta^{13}C = 0‰$ and $A^{14}C = 0$ pMC) but not due to radioactive decay. The location of experimental points in zone III may indicate arrival of very "young" ^{14}C even related to the "nuclear epoch" (normally tritium-bearing groundwater). The location of experimental points in zone I may be indicative of ^{14}C radioactive decay and validity of Equation 3 and also of arrival of the carbon with light isotopic content.

Considering the variability of α values, it is evident that for ages greater than 10–20 yr, calculation results slightly depend on the α value. From the ^{3}H data presented in Table 1, only 2 samples (S9 and C-2) are somewhat variable in their α coefficient. Residence times should be treated as preliminary and rather semiquantitatively, as they are based only on 1 sampling campaign performed so far. In general, having one ^{3}H measurement, only 1 parameter can be determined (normally groundwater residence time). However, the type of the model (piston model, PM; exponential model, EM; or dispersion model, DM) and additional parameters remain unknown. In order to determine the type of model and additional parameters using tracer data only, either a time series or additional tracers are required.

As well as groundwater residence time, tritium data give the additional possibility to determine the geochemical dilution factor of ^{14}C activity in the groundwater using ^{3}H versus ^{14}C activity and applying simple statistics (Figure 5).

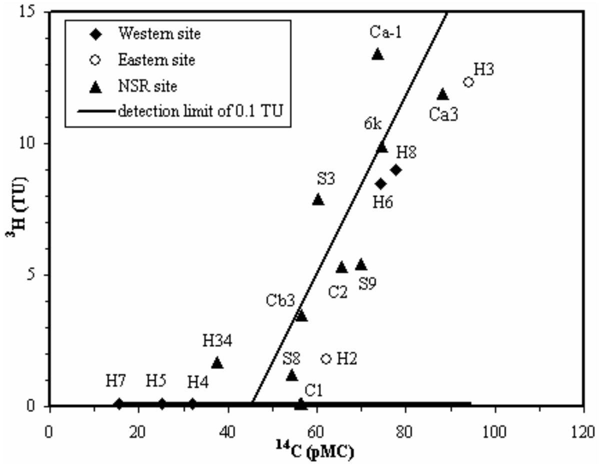

Figure 5 Relationship between ^{14}C and ^{3}H content of studied groundwaters. The initial ^{14}C activity was set to where the ^{14}C–^{3}H distribution curve hit the detection limit of ^{3}H. Statistically initial ^{14}C activity was estimated as 45 pMC (or q factor value of 0.45).

The initial ^{14}C activity for different investigated aquifers can be seen from the intersection of the fitted straight line to analytical data and the detection limit of ^{3}H (Figure 5). This initial ^{14}C activity approaches 45 pMC. Thus, Equation 4 for groundwater age evaluation with a q factor value of 0.45 (45/100 pMC) for the Quaternary aquifers can be used:

$$t_{age} = 8267 \ln\left(\frac{q \cdot A_{100pMC}}{^{14}C_{meas}}\right) \tag{4}$$

The ^{14}C activity in atmospheric CO_2 for pre-nuclear time is assumed to be 100 pMC. According to ^{14}C data, most of the groundwater samples studied represent modern groundwater. Their residence time in the aquifer at a depth up to 30 m varies from modern up to 60 yr. However, some groundwater samples, attributed as "modern" according to ^{14}C data, have no ^{3}H (residence time >58 yr) and can be treated as late Holocene in age. ^{14}C age can be determined for groundwater in confined aquifers from boreholes H7 (age of 10,500 yr, screening depth 61–65 m), H5 (4500 yr, screening depth 84–88 m), H4 (6500 yr, screening depth 129–134 m), and H34 (1900 yr, screening depth 33.6–36.0 m). The groundwater from the boreholes discussed is thus not influenced by the significant mixing with the modern water.

CONCLUSION

Groundwater characterization supported by isotope techniques was applied for selection and characterization of the site for the new NPP in Lithuania, as well as for selection and characterization of the site for the near-surface repository site. These studies were all within the framework of the projects related to decommissioning of the INPP.

At alternative sites for the new NPP, 2 flow systems with different groundwater circulation features were distinguished. Groundwater residence time in the first flow system until 30 m depth based on ^{3}H data ranges from 30 yr to ~60 yr and more. Groundwater residence time in the second flow system (60–134 m depth interval) based on ^{14}C data is in the order of several thousand years and in some intervals can reach up to 10,500 yr. Traces of glacier meltwater or prior frozen (cryolite zone) groundwater were detected in this deeper flow system according to the stable isotope data. Groundwater residence time inversions were observed for particular depth intervals, which can be related to the lower hydraulic conductivity of the first flow system compared with the second, as well as to the variable permeability of aquitards confining the studied aquifers.

In the NSR site, only the shallow groundwater flow system to a depth of 35 m was studied. For the groundwater taken from the majority of boreholes, the residence time in the aquifer can be assessed by ^{3}H data and varies from 6 yr up to ~57 yr. The groundwater residence time based on ^{14}C data was confirmed for borehole H34 as 1900 yr, at a screening depth of 33.6–36.0 m. Available data support low groundwater flow in shallow subsurface and weak hydraulic connection with deeper occurring productive aquifers. In this context, the selected sites are considered suitable for their planned use.

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