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$^3\text{H}/^3\text{He}$ Dating of Anthropogenic Tritium in a Shallow Alluvial Aquifer at Paks Nuclear Power Plant, Hungary

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Abstract

The tritium–helium-3 ($^3\text{H}/^3\text{He}$) dating method was applied to quantify groundwater apparent ages and estimate the migration of anthropogenic tritium in the shallow alluvial aquifer surrounding the Paks Nuclear Power Plant (Hungary). Groundwater samples were collected from monitoring wells between 2013 and 2016 and analyzed for tritium and dissolved noble gases. The investigated aquifer consists mainly of highly permeable sand and gravel deposits hydraulically connected to the Danube River. Reference wells indicate apparent groundwater ages between 26 and 43 years, with an average apparent $^3\text{H}/^3\text{He}$ age of approximately 37 years. Wells located within the operational area of the power plant show apparent $^3\text{H}/^3\text{He}$ ages ranging from 1.3 to 14.1 years, reflecting the transport of tritium released during leakage events associated with damaged sewer pipelines between 2005 and 2007. The spatial distribution of apparent ages reveals heterogeneous groundwater flow paths, and highlights the influence of well-screen sampling on age interpretation. The paper demonstrates that anthropogenic tritium released from nuclear infrastructure can serve as an effective age dating method and improve conceptual models of flow dynamics in shallow alluvial aquifers.

Keywords: $^3\text{H}/^3\text{He}$ dating; anthropogenic tritium; groundwater travel time; noble gases; alluvial aquifer; nuclear power plant contamination



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1. Introduction

The Paks Nuclear Power Plant (PNPP) is located in central Hungary on the right side of the Danube River near Paks city. PNPP has operated four VVER-440/V-213 type pressurized water reactors (PWR) since 1987. During their normal operations, PWRs generate (among other radionuclides) tritium (^3H). In Hungary, the Decree No. 15/2001 (VI.6.) of the Ministry of the Environment regulates the permissible limits of the release of radioactive materials into the environment. Although routine tritium releases remained well below regulatory limits, recurring detections in shallow groundwater indicated the presence of a local subsurface tritium source requiring hydrogeological interpretation. The regulatory limits for annual tritium releases are 20 TBq to the atmosphere and 600 TBq to

surface waters. Radioactive discharges in Hungary are regulated by Act CXVI of 1996 on Atomic Energy and Decree No. 15/2001 (VI. 6.) of the Minister for Environment, under the supervision of the Hungarian Atomic Energy Authority (HAEA). Average annual tritium emission between 2013 and 2017 was 4.3 TBq into the atmosphere and 24.7 TBq into surface waters (Danube River), well below the limits [1–4]. However, from the beginning of the operation, sporadic uncontrolled releases of a relatively small amount of tritium into the shallow groundwater system have been detected.

The expected residence times of the shallow groundwater fall within the effective dating range of the $^3\text{H}/^3\text{He}$ method, which has proven useful in numerous environmental studies [5–9]. For instance, the impact of a contamination hazard on drinking water quality can be investigated by using groundwater age. Since it provides useful information about the physical and chemical characteristics of the groundwater systems, it can help to predict the fate and transport of contaminants. As the ^3H peak, which originated from the thermonuclear bomb testing performed in the early 1960s, slowly declines to the natural level, new anthropogenic sources could be involved in isotope hydrology studies. New sources could be nuclear facilities, such as nuclear power plants and nuclear waste repositories [4,10–13]. In addition, industrial processes, which generate ^3H in their production cycle (e.g., clock manufacturers), could also be involved locally [14,15]. In such settings, anthropogenic tritium may act not only as a contaminant indicator but also as a transient tracer of groundwater movement and travel time.

In this work, we present a new application of the $^3\text{H}/^3\text{He}$ dating method to estimate the age of tritium contamination resulting from an uncontrolled release into a shallow groundwater aquifer at the PNPP. Based on the scientific cooperation established between the PNPP, the HUN-REN Institute for Nuclear Research, and Isotoptech Ltd., a four-year groundwater monitoring program was carried out between 2013 and 2016 within and around the operational area of the power plant. The groundwater monitoring system effectively identified potential source areas associated with historical tritium releases. The last significant tritium input to groundwater occurred between 2005 and 2007, when two sections of a sewer pipeline were damaged. During this period, maximum tritium activity concentrations approached 10^4 Bq L^{-1} (10^5 TU). Following the repair of both leakage points in 2007, tritium concentrations exhibited a fluctuating but generally declining trend. The use of anthropogenic (NPP-derived) tritium as a tracer for estimating apparent groundwater ages has only rarely been reported. Therefore, this study aimed to evaluate whether contamination from uncontrolled tritium releases can be used to improve the understanding of shallow groundwater dynamics and to estimate apparent groundwater ages in the study area.

Study Area

PNPP is situated in the middle of the Pannonian Basin, approximately 130 km south of capital of Hungary, Budapest. The facility is located 2 km from the Danube River at an elevation of about 97 m above sea level (m a.s.l.). The construction site was artificially raised above the Danube's flood level (Figure 1).

The region of the PNPP is an alluvial sedimentary area composed predominantly of sand and gravel deposits [16,17]. A local exception is the 3–5 m thick embankment, which consists of a heterogeneous mixture of silt, clay, and construction debris. The embankment material is poorly graded and characterized by low vertical hydraulic conductivity (10^{-4} – $10^{-1} \text{ m} \cdot \text{d}^{-1}$). Holocene fluvial deposits (sand with thin silt and fine gravel intercalations) and Pleistocene aeolian deposits (loess) occur beneath the embankment and reach a thickness of 20–30 m above the original ground surface [16–18]. The grain-size distribution generally becomes coarser with depth until the first continuous clayey aquitard, located

at a depth of approximately 30–40 m below ground level (Figure 2) [16]. The shallow unconfined alluvial aquifer is hydraulically connected to the Danube River, and hydraulic conductivity values determined from pumping tests conducted during site hydrogeological investigations range between 10^{-1} and 10^1 m·d $^{-1}$ [16,17].

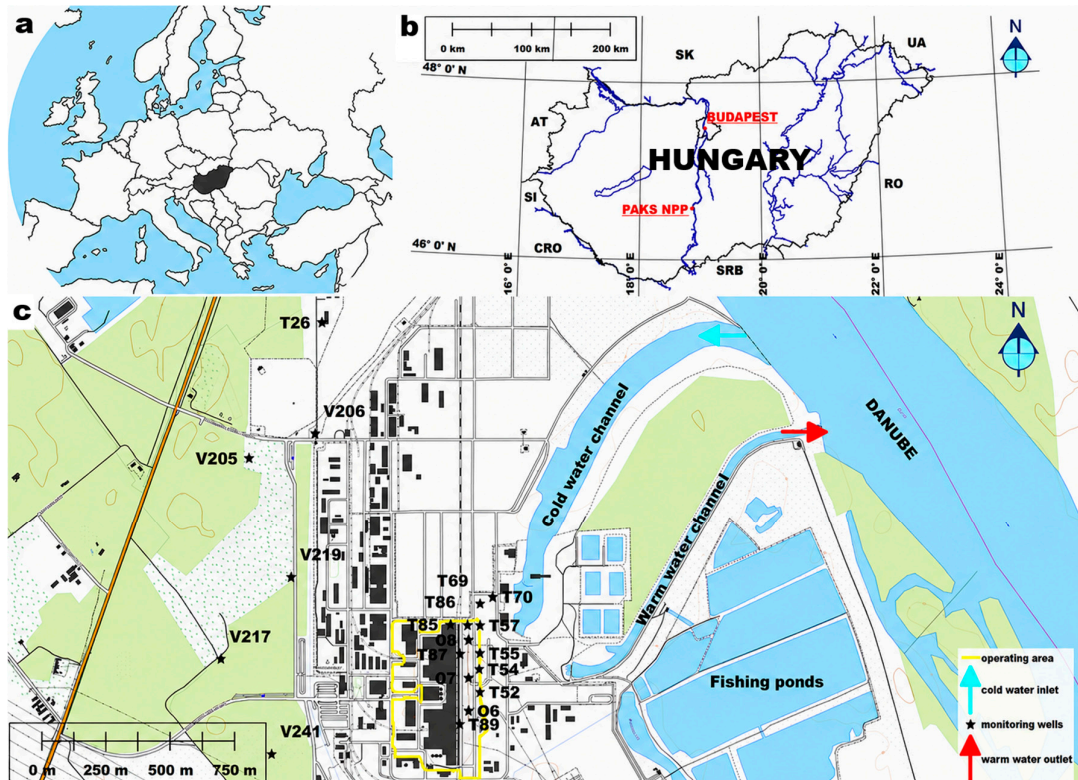


Figure 1. Location of Paks NPP in Europe (a) and Hungary (46.57° N, 18.85° E) (b), showing the sampled monitoring wells and the main surface water bodies (c).

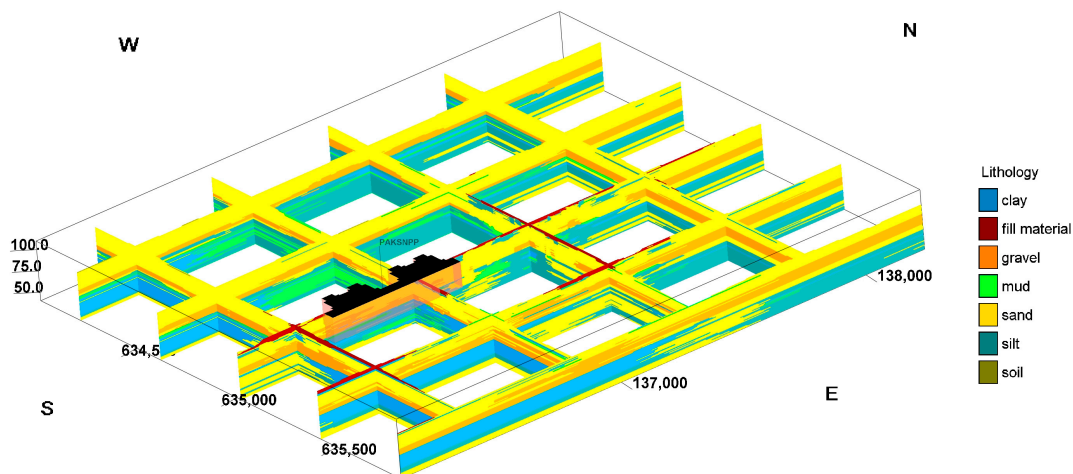


Figure 2. Lithological fence diagram of the immediate vicinity of the Paks Nuclear Power Plant (PNPP), showing elevations between 40 and 100 m a.s.l. The numbers shown along the figure margins represent map coordinates (m) in the Hungarian Unified National Projection (EOV) coordinate system.

The recharge area of the shallow aquifer is located on a loess plateau (140–180 m a.s.l.), 20 km west of the PNPP. The discharge area is the Danube, along with the Cold-Water Channel (CWCH). The hydraulic gradient for the shallow groundwater is ~ 2.5 m km $^{-1}$. The water table lies 8–10 m deep beneath the PNPP, at 87–89 m a.s.l. Based on the water

regime of the Danube in the last few decades, the water level usually ranges from 84 to 92 m a.s.l. The maximum annual water level fluctuation of the Danube approached 10 m in 2013. Groundwater levels and Danube water levels used in this study were obtained from the PNPP internal monitoring network, including monitoring wells and on-site water-level gauge measurements. Based on long-term observations, groundwater levels show a strong correlation with fluctuations in the Danube water level (Figure 3). Groundwater levels in the unconfined aquifer vary between 87 and 91 m a.s.l. in areas adjacent to surface water bodies and between 89 and 92 m a.s.l. west of the main plant buildings.

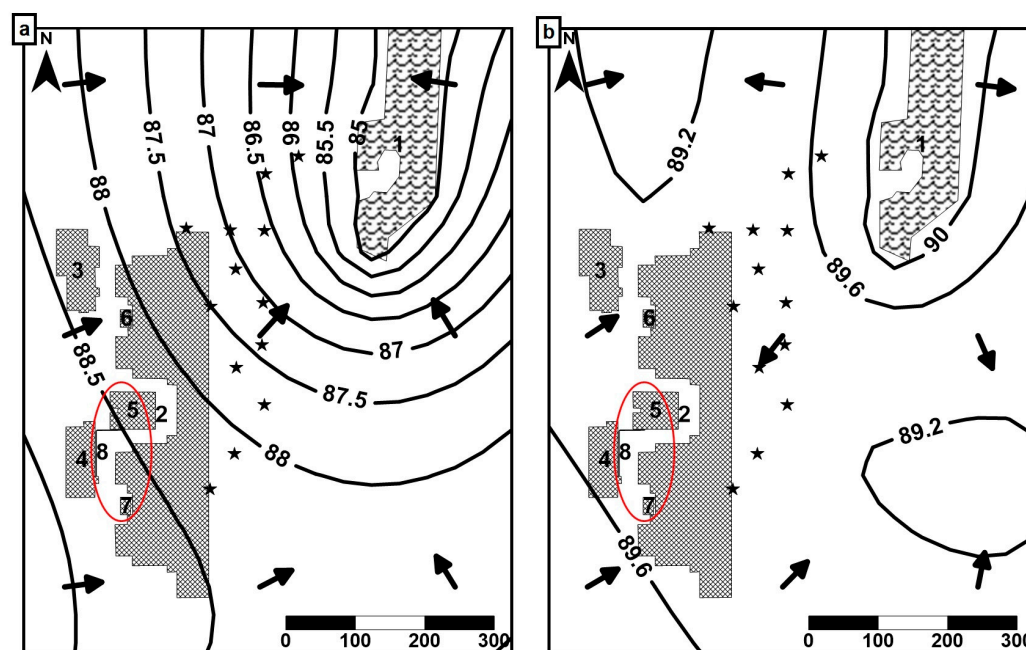


Figure 3. The hydraulic potential lines (m a.s.l.) and the horizontal flow direction of the local shallow groundwater table in case of low (a) and high (b) Danube levels. Black arrows indicate the inferred horizontal groundwater flow direction. 1. CWCH; 2. main building; 3., 4. Auxiliary buildings No. 1 and 2; 5. Sanitary facility and laboratory; 6., 7. water sumps; 8. low activity pipeline; red ellipse—potential contamination source. Data source: PNPP internal monitoring network (monitoring wells and on-site water-level gauge measurements).

The base of the main building extends into the saturated zone. The shallow groundwater table elevation is regulated by the water level of the Danube, and the dominant groundwater flow direction under the power plant is SW-NE, according to the local hydrodynamic model (Figure 3). From technological point of view, potential tritium sources could be found on the west side of the main building (Figure 3, red ellipse).

According to the Köppen climate classification, the studied area belongs to the warm temperate climate region [19], with an annual rainfall of 600–700 mm. The long-term average precipitation distribution is uneven and can be characterized by an early-summer maximum, as also reflected in the temporal variations in groundwater levels and surface water levels (Figure 4).

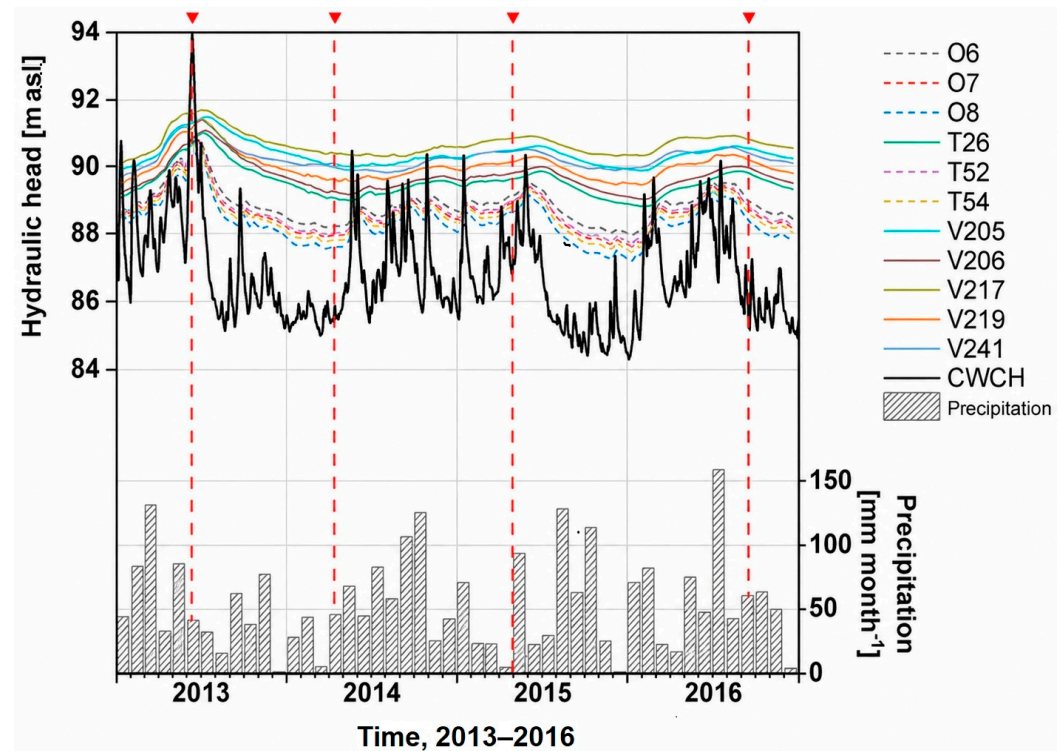


Figure 4. Relationship of the hydraulic heads measured in the sampled monitoring wells with the water levels of the Cold-Water Channel and the distribution of precipitation between 2013 and 2016. Red triangles show sampling events. Straight lines show reference wells, dotted lines show contaminated wells.

2. Materials and Methods

2.1. Sampling

The groundwater monitoring system of PNPP currently operates 114 wells drilled into the shallow aquifer with a maximum depth of ~25 m, and 2 wells drilled below the first continuous aquitard with a maximum depth of ~50 m. Hydraulic head is monitored in almost every well while tritium activity concentration is regularly measured in 50 wells in the close vicinity of the main building.

Based on the dominant groundwater flow direction, 19 monitoring wells were selected for the study. The well construction parameters and sampling depths are shown in Table 1. Sampling positions are reported both as elevation (m a.s.l.) and as depth below ground level (m b.g.l.), where depths were estimated as the difference between ground surface elevation and sampling elevation. To characterize the natural background conditions of the shallow groundwater system, six monitoring wells (T26, V205, V206, V217, V219, and V241) located more than 500 m west of the NPP were selected. As these wells are situated upgradient of the identified tritium source areas and consistently exhibit background tritium concentrations, they were used as uncontaminated reference wells. Additionally, 13 wells were chosen on the eastern side of the power plant. These wells (O6, O7, O8, T52, T54, T55, T57, T69, T70, T85, T86, T87, and T89) are located within the operational area of the NPP and along the expected migration pathways from the identified and suspected tritium source areas based on the local groundwater flow model. Therefore, they were considered potentially contaminated wells affected by NPP-related tritium releases.

To follow the tritium emission and distribution in the groundwater, a monitoring campaign was started in 2013. The groundwater samples were taken by a 2.5" pipe pump (IBO, DAMBAT Jastrzębski S.K.A., Adamów, Poland). The static water level was measured with a Heron dipper-T (Heron Instruments Inc., Dundas, ON, Canada) water level meter

before and after sampling. During the sampling, the following protocol was followed. Three well volumes of water were removed prior to sampling at a rate of 60 L min^{-1} following standard groundwater sampling protocols to obtain representative groundwater samples [20]. After purging, the rate of pumping was decreased to 1 L/min . Water samples were stored in 2 L polyethylene bottles for tritium analysis. At the same time, $\sim 40 \text{ cm}^3$ of water samples were taken into a copper tube sealed with pinched-off clamps for noble gas analysis. Groundwater samples were collected during four monitoring campaigns between 2013 and 2016 (11 June 2013, 27 February–6 March 2014, 28 April 2015, and 15 September–28 October 2016). Most monitoring wells were sampled once during each campaign, resulting in four samples per well. Additional depth-specific samples were collected from selected wells using the packer-based sampling system in 2016. Sampling was performed from the least contaminated wells to the most contaminated ones, to prevent cross-contamination.

For the first three years, groundwater samples were taken from a depth of 11 m, sampling the entire screened layer. A limitation of this approach is the relatively long screened intervals (5–15 m), which result in the collection of vertically integrated groundwater samples. To avoid collecting vertically averaged water samples, a special pump was developed in 2016. The sampler works similarly to a double packer system described by Zlotnik and McGuire (1998) [21]. Two inflatable rubber hoses (packers) enclose three pumps connected serially. Both can be inflated from the wellhead with a compressor. In this way, a one-meter section was separated along the well screen. The three small-capacity pumps lifted the groundwater to the surface for sampling (Figure 5). Packer-based depth-specific sampling was performed in six monitoring wells (O6, O7, O8, T57, T69, and T70) during the 2016 sampling campaigns (September–October 2016). In these wells, hydraulic conditions allowed the isolation of a 1 m section within the screened interval.

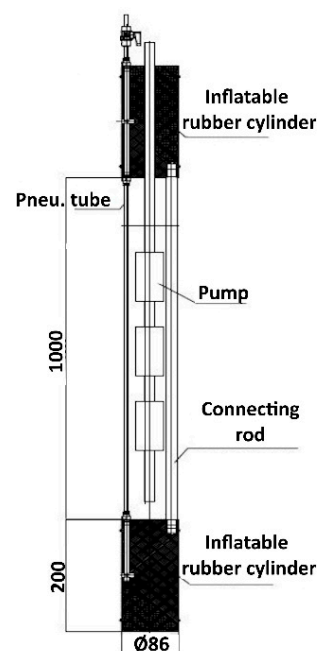


Figure 5. Schematic design of the specialized double packer sampling system. The device features two inflatable rubber cylinders (packers) that isolate a 1 m vertical section within the well screen. Three serially connected low-capacity pumps are positioned between the packers to extract groundwater samples from the isolated interval, preventing vertical averaging.

Table 1. Construction parameters of the monitoring wells and applied sampling intervals. Elevations are given in m a.s.l., depths in m b.g.l.; n.d.—no data.

Well ID	Packer Sampling (2016)	Surface (m a.s.l.)	Sampling Elevation (m a.s.l.)				Sampling Depth (m b.g.l.)							
			Screen		Sampling Interval 2013–2015		Sampling Interval 2016		Screen Depth		Sampling Interval 2013–2015		Sampling Interval 2016	
			Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom
O6	yes	97.1	91.1	83.1	86	85.5	89	88	6	14	11.1	11.6	8.1	9.1
							85	84					12.1	13.1
O7	yes	97	91	83	86	85.5	89	88	6	14	11	11.5	8	9
							85	84					12	13
O8	yes	97.2	91.2	83.2	86	85.5	88	87	6	14	11.2	11.7	9.2	10.2
							85	84					12.2	13.2
T26	no	97	88.5	83.5	86	85.5	86	85.5	8.5	13.6	11	11.5	11	11.5
T52	no	97.2	89.2	84.2	86	85.5	86	85.5	8	12.9	11.2	11.7	11.2	11.7
T54	no	96.8	88.8	83.8	86	85.5	86	85.5	8	13.3	10.8	11.3	10.8	11.3
T55	no	96.9	81.9	78.9	80.5	80	80.5	80	15	18.2	16.4	16.9	16.4	16.9
T57	yes	97.2	94.7	79.2	86	85.5	87	86	2.5	17.9	11.2	11.7	10.2	11.2
							81	80					16.2	17.2
T69	yes	97.1	94.6	79.1	86	85.5	87	86	2.5	18	11.1	11.6	10.1	11.1
							82	81					15.1	16.1
T70	yes	96.9	94.4	78.9	86	85.5	86.5	85.5	2.5	18.2	10.9	11.4	10.4	11.4
							82	81					14.9	15.9
T85	no	96.9	92.6	84.6	86	85.5	86	85.5	4.3	12.5	10.9	11.4	10.9	11.4
T86	no	97	92.8	84.8	86	85.5	86	85.5	4.2	12.3	11	11.5	11	11.5
T87	no	96.9	93.3	85.3	86	85.5	86	85.5	3.6	11.8	10.9	11.4	10.9	11.4
T89	no	97	93	85	86	85.5	86	85.5	4	12.1	11	11.5	11	11.5
V205	no	95.3	n.d.	n.d.	86	85.5	86	85.5	n.d.	n.d.	9.3	9.8	9.3	9.8
V206	no	97.1	87.1	82.1	86	85.5	86	85.5	10	15	11.1	11.6	11.1	11.6

Table 1. Cont.

Well ID	Packer Sampling (2016)	Surface (m a.s.l.)	Sampling Elevation (m a.s.l.)						Sampling Depth (m b.g.l.)					
			Screen		Sampling Interval 2013–2015		Sampling Interval 2016		Screen Depth		Sampling Interval 2013–2015		Sampling Interval 2016	
			Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom
V217	no	96.6	86.6	81.6	86	85.5	86	85.5	10	15.5	10.6	11.1	10.6	11.1
V219	no	94.6	n.d.	n.d.	86	85.5	86	85.5	n.d.	n.d.	8.6	9.1	8.6	9.1
V241	no	95.2	85.6	80.6	85	84.5	85	84.5	9.6	16.5	10.2	10.7	10.2	10.7

2.2. Analytical Methods

The analytical method was selected according to the expected tritium concentration range derived from routine site-monitoring data collected prior to this study. Samples with low tritium concentration (non-contaminated natural groundwater) were analyzed by the ^3He ingrowth method [22–24], and high-concentration samples by liquid scintillation counting. Tritium and noble gas measurements were carried out using a TriCarb3180 TR/SL (PerkinElmer, Waltham, MA, USA), and Quantulus 1220 liquid scintillation counters (Perkin-Elmer, Waltham, MA, USA) and a VG5400 (Fisons Instruments, Winsford, UK now Thermo Scientific) noble gas mass spectrometer. Tritium as well as dissolved ^3He are expressed in tritium units (TU), where 1 TU represents a $^3\text{H}/\text{H}$ or $^3\text{He}/\text{H}$ ratio of 10^{-18} . In the case of water, 1 TU equals 0.119 Bq/kg activity concentration. In case of the tritiogenic ^3He dissolved in water, 1 TU equals 2.49×10^{-15} ccSTP/g of dissolved ^3He (ccSTP: cubic centimeter at standard temperature and pressure). The detection limit for tritium analysis was better than 0.012 TU. The relative analytical uncertainty was typically 2–4% (1σ) in the concentration range of 1–20 TU and 1–5% above 20 TU. Noble gas measurements were calibrated against air standards of known composition, yielding analytical precisions of 1.0% for helium and argon and 1.5% for neon, krypton, and xenon.

2.3. Calculations

The tritium (^3H) has a half-life ($T_{1/2}$) of 12.32 ± 0.02 years [25], making it an essential tool for dating young groundwater (between 0 and 70 years). Taking advantage of its short half-life and using it with its decay product (^3He), tritium gives the opportunity to calculate an apparent age for modern groundwater. The theoretical background of the method has been described in detail by several authors [5,6,26–36]. In this context, “age” should be referred to as the time elapsed since a water body has been isolated from interacting with atmospheric gases.

As the recharged water is not in contact with the atmosphere any longer, the dissolved ^3He ($^3\text{He}_{\text{water}}$) can be decomposed into these components:

$$^3\text{He}_{\text{water}} = ^3\text{He}_{\text{eq}} + ^3\text{He}_{\text{ex}} + ^3\text{He}_{\text{terr}} + ^3\text{He}_{\text{trit}} + ^3\text{He}_{\text{trit,NPP}}$$

where the $^3\text{He}_{\text{eq}}$ is the solubility equilibrium, the $^3\text{He}_{\text{ex}}$ is the excess air and the $^3\text{He}_{\text{terr}}$ is the terrigenous component. The different components can be calculated by means of the recharge temperature as well as the other atmospheric noble gas concentrations (Ne, Ar, Kr and Xe) [37,38].

The R/Ra ratio is the measured $^3\text{He}/^4\text{He}$ (R) and the air $^3\text{He}/^4\text{He}$ ratio ($\text{Ra} = 1.384 \times 10^{-6}$). When R/Ra is higher than 1, there is a clear indication of either $^3\text{He}_{\text{trit}}$ (natural and bomb) or $^3\text{He}_{\text{trit,NPP}}$.

The tritiogenic helium component ($^3\text{He}_{\text{trit}}$) originates from the radioactive decay of natural and bomb-derived tritium recharged with precipitation. This tritium recharges into the subsurface with the precipitation. In this study, a separate component ($^3\text{He}_{\text{trit,NPP}}$) was defined to represent tritiogenic helium produced by the decay of anthropogenic tritium released from the nuclear power plant. Usually, the $^3\text{He}_{\text{trit}}$ component is used for $^3\text{H}/^3\text{He}$ age determination in isotope hydrology [5,8,27,39,40]. In this study, background $^3\text{He}_{\text{trit}}$ values were determined using samples from unaffected, uncontaminated groundwater bodies far enough away and counter flow from the NPP. In addition, a separate component $^3\text{He}_{\text{trit,NPP}}$ was defined to account for helium coming from the decay of the unintended tritium release from the PNPP.

The apparent age (τ) was calculated using the $^3\text{H}/^3\text{He}$ method as:

$$\tau = T_{1/2}/\ln 2 \times \ln \left(1 + \left[^3\text{He}_{\text{trit,NPP}} \right] / \left[^3\text{H} \right] \right)$$

where τ is the apparent age, ^3H is the measured activity concentration, and $^3\text{He}_{\text{trit,NPP}}$ is tritiogenic helium attributed to the NPP-derived tritium.

This apparent age calculation assumes (i) that the system is closed, i.e., ^3He is not released from the groundwater, and (ii) zero initial tritiogenic helium is associated with the released tritium at the time of entry into the subsurface. Uncertainties are reported as ± 1 standard deviation (SD).

In addition, lumped parameter modeling (LPM) of the uncontaminated wells was performed using TracerLPM (U.S. Geological Survey, Reston, VA, USA) [41] and Lumpy [24,39] with ^3H and $^3\text{He}_{\text{trit}}$ as input tracers. Detailed model descriptions and parameterization are provided in the Supplementary Materials.

2.4. Limitations of the Sampling Approach

Only six wells met the hydraulic conditions required for packer-based depth-specific sampling due to groundwater level limitations. In the remaining wells, the water table did not reach the top of the screened interval.

Consequently, samples from these wells represent vertically integrated water over relatively long screened sections (5–15 m), which may lead to mixing of groundwater of different ages. These limitations are taken into account in the interpretation of the results.

3. Results

3.1. Reference Wells

Assuming a sampling depth of 11 m in the reference wells and 8 ± 1 m near the NPP, the bomb contribution can be estimated. The measured tritium concentrations in the reference wells range between ~3 and 17 TU (Table 2), consistent with low and relatively stable background levels. Figure 6 shows the initial tritium concentration ($^3\text{H}_{\text{initial}}$) as a function of the recharge calendar year. The recharge years were calculated from the $^3\text{H}/^3\text{He}$ apparent ages and the corresponding sampling dates.

The average of these calendar years is 1977 (November) ± 4.2 years.

Assuming a constant recharge rate and vertical groundwater flow, at a depth of 8 m, water ages near the nuclear power plant are 10 years younger (calendar year: 1987) than those in reference wells at 11 m depth. Here, the initial tritium concentration is 20.0 ± 13.2 TU.

Hence, the bomb contribution at 8 m is then the difference between $^3\text{H}_{\text{initial}}$ (in 1987) and $^3\text{H}_{\text{measured,average}}$ (in 2013–2016). The average tritium concentration of the reference wells is 4.4 ± 3.2 TU, so the $^3\text{H}_{\text{bomb}}$ (at 8 m) equals 15.6 ± 13.6 TU.

In the six uncontaminated wells (T26, V205, V206, V217, V219, V241), the average apparent age of the groundwater is 37 ± 4.3 years without the highlighted data. Including all the measured data, this result is 35.4 ± 8.7 years. The highlighted results were excluded from the calculation because their isotope ratios and apparent ages significantly deviated from the regional trend observed in the uncontaminated monitoring wells, suggesting potential analytical uncertainty or local-scale hydrogeological effects.

The R/Ra ratios of reference wells vary around 2.8 ± 0.5 . The $^3\text{H}_{\text{initial}}$ values in reference wells range between 45 and 86 TU. A mean value of 60.7 ± 12.0 TU was obtained for $^3\text{H}_{\text{initial}}$ after excluding outliers (Table 2).

These results characterize the natural baseline conditions of the aquifer in terms of tritium content, helium isotope composition, and groundwater residence time.

Table 2. The noble gas and age results of the uncontaminated monitoring wells. R/R_a represents the ratio between ³He/⁴He isotopes in the water samples (R) and in the atmosphere (R_a). Highlighted results are excluded from the calculation. * Sample was remeasured.

Well ID	Sampling Date	He (10 ^{−8} ccSTP/g)	Ne (10 ^{−7} ccSTP/g)	Ar (10 ^{−4} ccSTP/g)	Kr (10 ^{−8} ccSTP/g)	Xe (10 ^{−8} ccSTP/g)	R/R _a	³ H (TU)	Unc. (TU)	³ He _{trit} (TU)	Unc. (TU)	³ H/ ³ He Age (Year)	Unc. (Year)
V205	11 June 2013	5.21 × 10 ^{−8}	2.19 × 10 ^{−7}	3.77 × 10 ^{−4}	8.45 × 10 ^{−8}	1.17 × 10 ^{−8}	3.01	6.52	0.16	58.31	2.21	40.8	0.7
	27 February 2014	5.26 × 10 ^{−8}	1.78 × 10 ^{−7}	3.86 × 10 ^{−4}	8.47 × 10 ^{−8}	1.24 × 10 ^{−8}	3.23	7.15	0.19	63.1	2.4	40.6	0.7
	27 February 2014 *	5.33 × 10 ^{−8}	2.16 × 10 ^{−7}	3.79 × 10 ^{−4}	8.63 × 10 ^{−8}	1.24 × 10 ^{−8}	3.06	7.15	0.19	62.75	2.3	40.5	0.7
	28 April 2015	6.45 × 10 ^{−8}	2.43 × 10 ^{−7}	3.78 × 10 ^{−4}	8.81 × 10 ^{−8}	1.31 × 10 ^{−8}	3.23	6.43	0.24	90.99	2.93	48.3	0.8
	28 April 2015 *	5.62 × 10 ^{−8}	2.35 × 10 ^{−7}	3.75 × 10 ^{−4}	8.68 × 10 ^{−8}	1.32 × 10 ^{−8}	3.08	6.43	0.24	71.21	2.44	44.3	0.8
	28 October 2016	5.29 × 10 ^{−8}	2.31 × 10 ^{−7}	3.67 × 10 ^{−4}	8.28 × 10 ^{−8}	1.20 × 10 ^{−8}	3.58	7.77	0.14	78.3	2.66	42.7	0.6
V206	6 March 2014	4.52 × 10 ^{−8}	1.40 × 10 ^{−7}	3.81 × 10 ^{−4}	8.43 × 10 ^{−8}	1.24 × 10 ^{−8}	1.43	7.82	0.21	6.5	0.98	10.8	1.2
	28 April 2015	4.84 × 10 ^{−8}	1.88 × 10 ^{−7}	3.55 × 10 ^{−4}	8.64 × 10 ^{−8}	1.32 × 10 ^{−8}	2.29	8.39	0.31	36.69	1.59	29.9	0.8
	28 October 2016	4.92 × 10 ^{−8}	2.51 × 10 ^{−7}	3.60 × 10 ^{−4}	8.00 × 10 ^{−8}	1.12 × 10 ^{−8}	1.89	7.53	0.07	21.73	1.35	24.1	0.8
V217	11 June 2013	4.96 × 10 ^{−8}	2.10 × 10 ^{−7}	3.82 × 10 ^{−4}	8.53 × 10 ^{−8}	1.22 × 10 ^{−8}	2.54	8.52	0.21	42.7	1.79	31.9	0.7
	27 February 2014	4.74 × 10 ^{−8}	2.13 × 10 ^{−7}	3.84 × 10 ^{−4}	8.42 × 10 ^{−8}	1.20 × 10 ^{−8}	2.91	8.65	0.23	43.19	1.96	31.8	0.8
	27 February 2014 *	5.08 × 10 ^{−8}	1.87 × 10 ^{−7}	3.79 × 10 ^{−4}	8.79 × 10 ^{−8}	1.27 × 10 ^{−8}	2.87	8.65	0.23	54.1	2.06	35.2	0.7
	28 April 2015	5.28 × 10 ^{−8}	2.23 × 10 ^{−7}	3.75 × 10 ^{−4}	8.80 × 10 ^{−8}	1.33 × 10 ^{−8}	2.96	7.83	0.29	61.93	2.21	38.9	0.8
	28 October 2016	4.94 × 10 ^{−8}	2.07 × 10 ^{−7}	3.80 × 10 ^{−4}	8.71 × 10 ^{−8}	1.23 × 10 ^{−8}	2.64	6.18	0.06	45.8	1.85	37.9	0.6
V219	11 June 2013	5.25 × 10 ^{−8}	2.14 × 10 ^{−7}	3.86 × 10 ^{−4}	8.68 × 10 ^{−8}	1.23 × 10 ^{−8}	2.5	6.56	0.16	45.29	1.87	36.7	0.7
	27 February 2014	4.72 × 10 ^{−8}	2.14 × 10 ^{−7}	3.83 × 10 ^{−4}	8.52 × 10 ^{−8}	1.23 × 10 ^{−8}	2.65	6.65	0.18	41.66	1.78	35.3	0.8
	6 March 2014	4.95 × 10 ^{−8}	2.28 × 10 ^{−7}	3.91 × 10 ^{−4}	8.68 × 10 ^{−8}	1.24 × 10 ^{−8}	2.69	6.35	0.17	44.08	1.89	36.8	0.8
	28 April 2015	5.86 × 10 ^{−8}	2.38 × 10 ^{−7}	3.81 × 10 ^{−4}	8.77 × 10 ^{−8}	1.31 × 10 ^{−8}	2.68	5.99	0.22	62.33	2.22	43.3	0.8
	28 October 2016	5.16 × 10 ^{−8}	2.13 × 10 ^{−7}	3.85 × 10 ^{−4}	8.76 × 10 ^{−8}	1.25 × 10 ^{−8}	2.4	5.43	0.05	41.32	1.76	38.3	0.7
V241	27 February 2014	4.78 × 10 ^{−8}	1.89 × 10 ^{−7}	3.78 × 10 ^{−4}	8.41 × 10 ^{−8}	1.21 × 10 ^{−8}	3.39	11.26	0.33	59.8	2.29	32.7	0.7
	6 March 2014	4.41 × 10 ^{−8}	2.12 × 10 ^{−7}	3.77 × 10 ^{−4}	8.39 × 10 ^{−8}	1.20 × 10 ^{−8}	3.38	10.76	0.28	55.42	2.11	32.3	0.7
	28 April 2015	4.98 × 10 ^{−8}	2.20 × 10 ^{−7}	3.66 × 10 ^{−4}	8.50 × 10 ^{−8}	1.25 × 10 ^{−8}	3.12	3.18	0.12	61.59	2.2	53.6	0.9
	28 October 2016	4.87 × 10 ^{−8}	2.25 × 10 ^{−7}	3.72 × 10 ^{−4}	8.46 × 10 ^{−8}	1.20 × 10 ^{−8}	2.62	7.27	0.09	41.02	1.81	33.6	0.7
T26	6 March 2014	4.22 × 10 ^{−8}	1.94 × 10 ^{−7}	3.63 × 10 ^{−4}	8.19 × 10 ^{−8}	1.19 × 10 ^{−8}	3.3	16.67	0.46	52.69	1.97	25.3	0.6
	28 April 2015	5.39 × 10 ^{−8}	1.95 × 10 ^{−7}	3.59 × 10 ^{−4}	8.56 × 10 ^{−8}	1.30 × 10 ^{−8}	3.27	17.08	0.62	73.24	2.48	29.6	0.7
	28 October 2016	3.71 × 10 ^{−8}	1.71 × 10 ^{−7}	3.09 × 10 ^{−4}	7.36 × 10 ^{−8}	1.08 × 10 ^{−8}	3.22	15.3	0.13	44.87	1.7	24.3	0.5

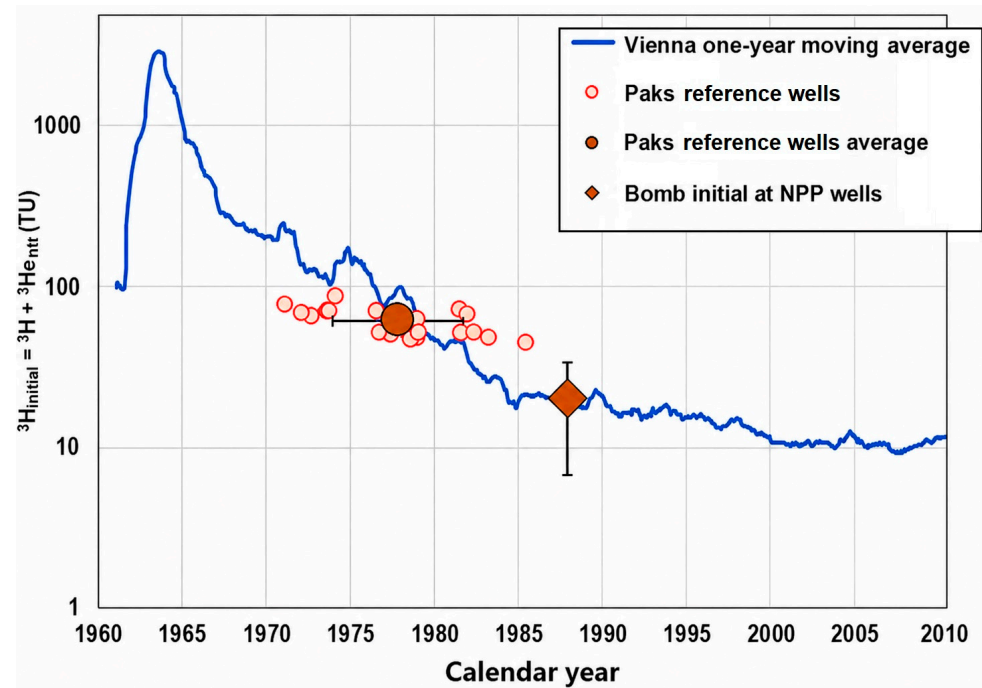


Figure 6. The calculated $^3\text{H}_{\text{initial}}$ ($=^3\text{H}_{\text{measured}} + ^3\text{He}_{\text{trit}}$) concentration of the reference wells in function of the calendar years of recharge. The values of the reference wells (small circles) vary in the range of 60.7 ± 12.0 around the calendar date of November 1977 ± 4.2 years (big circle). The initial tritium concentration of the recharge water at 8 m depth is obtained to be 20.0 ± 13.2 TU (big diamond).

3.2. Contaminated Wells

Tritium concentrations in the contaminated wells range from tens to over 10^3 TU (Table 3), allowing classification into moderately (10–100 TU, e.g., O6/1, O7/1, T70/2, T85/2016), highly (100–500 TU, e.g., O6, O8/1, O8/2, T54, T57, T69/2) and extremely contaminated groups (>500 TU, e.g., O7, O8, T52, T55, T69, T70, T86). A clear excess of tritogenic ^3He originating from the NPP is observed in the samples taken from the operating area.

Measured noble gas concentrations (He, Ne, Ar, Kr, Xe) in the contaminated wells show a wider range of values compared to the reference wells (Table 3). For example, helium concentrations range from ~ 3.6 to 6.6×10^{-8} ccSTP/g, while neon concentrations vary between ~ 1.5 and 3.2×10^{-7} ccSTP/g (Table 3). The R/Ra values of the monitoring wells vary between 1.7 and 34.7, indicating a remarkable excess of ^3He . For results with a low R/Ra ratio, the bomb-peak correction has a considerable effect (Table 3).

The apparent ages range between 1.3 ± 2.1 and 14.1 ± 0.4 years, with most values falling between ~ 4 and 10 years (Table 3).

Depth-specific sampling with the packer sampling unit revealed differences relative to bulk samples from long-screened wells, indicating that vertical mixing can significantly affect apparent ages. In some cases, the packer-derived samples showed more distinct age signals across isolated depth intervals within the same well (e.g., O6, T69). For example, apparent ages in well O6 ranged from 1.3 ± 2.1 to 6.0 ± 1.3 years within isolated intervals, while in well T69 values between 10.8 ± 1.0 and 11.7 ± 2.3 years were observed.

These results indicate the presence of a young and spatially variable tritium signal in the operational area.

Table 3. The noble gas and age results of the contaminated monitoring wells.

Well ID	Sampling Date	He (10^{-8} ccSTP/g)	Ne (10^{-7} ccSTP/g)	R/Ra	^3H (TU)	Unc. (TU)	$^3\text{He}_{\text{trit}}$ (TU)	Unc. (TU)	$^3\text{H}/^3\text{He}$ Age (Year)	Unc. (Year)
O6	11 June 2013	5.827×10^{-8}	2.369×10^{-7}	6.21	198.4	4.0	163.36	5.12	10.2	0.7
	14 April 2014	5.371×10^{-8}	2.229×10^{-7}	2.61	115.8	3.1	42.39	2.15	4.5	1.5
	27 April 2015	5.969×10^{-8}	2.699×10^{-7}	3.33	133.2	5.1	67.35	2.97	6.4	1.2
O6/1	15 September 2016	6.551×10^{-8}	2.830×10^{-7}	1.69	96.17	5.44	16.22	1.91	1.3	2.1
O6/2	15 September 2016	5.252×10^{-8}	2.340×10^{-7}	3.29	123.67	5.80	58.64	2.58	6.0	1.3
O7	11 June 2013	5.043×10^{-8}	2.146×10^{-7}	16.79	1078.7	21.6	436.72	11.81	5.9	0.2
	14 April 2014	4.964×10^{-8}	1.752×10^{-7}	18.78	1091.3	14.9	490.33	12.98	6.5	0.2
	27 April 2015	6.570×10^{-8}	3.179×10^{-7}	7.21	266.9	6.7	212.62	6.71	10.1	0.6
O7/1	15 September 2016	5.298×10^{-8}	2.358×10^{-7}	2.10	27.35	4.15	24.21	1.82	7.8	4.4
O7/2	15 September 2016	5.606×10^{-8}	2.294×10^{-7}	11.77	418.22	9.77	330.42	9.23	10.1	0.4
O8	11 June 2013	5.043×10^{-8}	2.146×10^{-7}	15.67	794.9	15.9	405.32	11.03	7.2	0.3
	14 April 2014	5.110×10^{-8}	2.054×10^{-7}	23.53	1605.4	20.2	635.97	16.74	5.9	0.2
	27 April 2015	4.832×10^{-8}	2.035×10^{-7}	14.75	691.6	12.0	363.72	9.94	7.4	0.3
O8/1	15 September 2016	4.386×10^{-8}	1.909×10^{-7}	10.13	341.23	8.98	216.47	6.22	8.4	0.5
O8/2	15 September 2016	3.557×10^{-8}	1.518×10^{-7}	11.25	393.59	9.94	198.31	5.60	7.0	0.5
T52	11 June 2013	5.845×10^{-8}	2.192×10^{-7}	7.97	893.7	17.9	223.85	6.53	3.8	0.2
	14 April 2014	5.424×10^{-8}	2.310×10^{-7}	7.11	233.7	6.2	177.49	5.44	9.6	0.7
	27 April 2015	5.138×10^{-8}	2.134×10^{-7}	6.95	173.0	5.9	164.47	5.04	11.4	0.8
	15 September 2016	5.010×10^{-8}	2.168×10^{-7}	3.16	134.55	6.11	53.44	2.37	5.1	1.3
T54	11 June 2013	6.367×10^{-8}	2.167×10^{-7}	3.69	221.3	4.4	96.11	3.38	5.9	0.8
	14 April 2014	5.162×10^{-8}	2.150×10^{-7}	6.24	192.7	5.6	144.84	4.56	9.5	0.8
	27 April 2015	4.871×10^{-8}	2.388×10^{-7}	9.43	381.8	8.4	217.14	6.46	7.7	0.5
	15 September 2016	5.281×10^{-8}	2.332×10^{-7}	6.79	209.58	7.43	162.09	5.07	9.7	0.7
T55	11 June 2013	5.043×10^{-8}	2.146×10^{-7}	9.92	1541.3	30.8	243.87	7.01	2.5	0.2
	14 April 2014	5.245×10^{-8}	1.872×10^{-7}	23.30	1019.9	14.4	649.85	17.01	8.7	0.3
	27 April 2015	5.540×10^{-8}	2.363×10^{-7}	34.74	1432.0	19.2	1032.70	26.78	9.6	0.2
	15 September 2016	4.228×10^{-8}	1.978×10^{-7}	16.11	723.22	13.30	347.07	9.50	6.8	0.3
T57	11 June 2013	5.043×10^{-8}	2.146×10^{-7}	7.94	617.1	12.3	188.48	5.64	4.5	0.3
	14 April 2014	4.493×10^{-8}	1.963×10^{-7}	14.78	742.9	11.5	337.97	9.27	6.5	0.3
	27 April 2015	4.123×10^{-8}	1.966×10^{-7}	10.53	742.3	12.3	209.88	6.09	4.3	0.3
T57/1	27 October 2016	4.745×10^{-8}	1.210×10^{-7}	9.10	526.33	10.70	220.73	6.02	6.0	0.4
T57/2	27 October 2016	4.945×10^{-8}	2.296×10^{-7}	8.26	472.99	10.05	190.42	5.75	5.8	0.4
T69	11 June 2013	5.972×10^{-8}	1.823×10^{-7}	21.73	946.8	18.9	692.68	18.06	9.7	0.3
	14 April 2014	5.025×10^{-8}	2.253×10^{-7}	26.35	1208.0	29.2	700.45	18.44	8.0	0.3
	27 April 2015	4.926×10^{-8}	2.468×10^{-7}	11.18	370.6	8.6	266.70	7.72	9.4	0.5
T69/1	15 September 2016	5.090×10^{-8}	2.338×10^{-7}	3.38	52.60	4.81	58.32	2.57	11.7	2.3
T69/2	27 October 2016	4.666×10^{-8}	2.028×10^{-7}	6.34	146.99	5.91	132.15	4.20	10.8	1.0
T70	11 June 2013	6.168×10^{-8}	2.171×10^{-7}	15.95	771.8	15.4	512.28	13.70	8.9	0.3
	14 April 2014	4.739×10^{-8}	1.716×10^{-7}	6.64	221.5	5.4	147.58	4.43	8.6	0.7
	27 April 2015	6.265×10^{-8}	2.871×10^{-7}	4.31	126.3	5.3	104.08	3.92	10.0	1.1
T70/1	27 October 2016	5.360×10^{-8}	2.020×10^{-7}	8.30	588.34	11.62	215.00	6.24	5.3	0.3
T70/2	27 October 2016	5.199×10^{-8}	1.783×10^{-7}	7.61	393.57	9.34	191.59	5.55	6.8	0.5
T85	11 June 2013	4.452×10^{-8}	1.841×10^{-7}	14.49	1006.8	20.1	329.36	9.00	4.9	0.2
	14 April 2014	4.769×10^{-8}	2.020×10^{-7}	5.51	290.7	6.5	113.82	3.74	5.5	0.6
	27 April 2015	5.533×10^{-8}	1.678×10^{-7}	6.18	239.9	6.6	163.19	4.80	8.8	0.7
	15 September 2016	4.989×10^{-8}	2.363×10^{-7}	1.88	31.97	4.27	14.18	1.61	2.6	4.7
T86	11 June 2013	5.043×10^{-8}	2.146×10^{-7}	25.68	560.2	11.2	686.18	18.03	14.1	0.4
	14 April 2014	4.302×10^{-8}	1.836×10^{-7}	24.62	827.1	12.8	559.73	14.75	9.1	0.3
	27 April 2015	5.664×10^{-8}	2.273×10^{-7}	16.46	881.5	13.6	482.32	13.00	7.6	0.3
	15 September 2016	4.579×10^{-8}	2.235×10^{-7}	13.72	307.99	8.04	313.77	8.79	12.2	0.5
T87	11 June 2013	5.800×10^{-8}	2.252×10^{-7}	5.07	366.7	7.3	127.60	4.19	5.0	0.5
	14 April 2014	4.950×10^{-8}	1.851×10^{-7}	5.67	160.7	5.1	126.49	3.97	9.7	0.9
	27 April 2015	5.579×10^{-8}	2.248×10^{-7}	4.42	281.0	7.1	101.11	3.54	5.0	0.7
	15 September 2016	5.169×10^{-8}	2.273×10^{-7}	5.49	374.33	10.03	121.57	4.05	4.7	0.5
T89	11 June 2013	5.979×10^{-8}	2.344×10^{-7}	1.96	112.7	2.3	27.46	1.88	2.7	1.7
	14 April 2014	5.388×10^{-8}	1.911×10^{-7}	2.45	150.5	5.2	42.75	1.99	3.6	1.2
	27 April 2015	5.833×10^{-8}	2.399×10^{-7}	2.33	133.67	5.00	37.29	2.12	3.4	1.4
	15 September 2016	5.184×10^{-8}	2.385×10^{-7}	2.56	304.40	8.15	35.71	2.08	1.5	0.7

3.3. Tracer Relationships (^3H , Noble Gases, R/Ra , $^3\text{H}/^3\text{He}$ Ages)

The combined evaluation of tritium concentrations, noble gas compositions and helium isotope ratios allows a consistent distinction between natural and anthropogenic (NPP-derived) tritium influenced groundwater. Reference wells are characterized by low tritium concentrations (4.4 ± 3.2 TU), stable R/Ra ratios ($\sim 2.8 \pm 0.5$), and relatively old apparent ages (26–43 years). In contrast, contaminated wells show elevated tritium concentrations, significantly higher and more variable R/Ra ratios (1.7–34.7), and younger apparent ages (1.3–14.1 years).

The magnitude of the tritiogenic ^3He excess is positively correlated with tritium concentration, resulting in reduced uncertainties in apparent age calculations in contaminated wells.

These systematic differences between reference and contaminated wells confirm the applicability of the combined tracer approach for identifying anthropogenic (NPP-derived) tritium influence and estimating apparent groundwater ages, as summarized in Table 4.

Table 4. Summary of tritium concentrations and $^3\text{H}/^3\text{He}$ apparent ages in reference and contaminated monitoring wells. Values represent characteristic ranges observed during the 2013–2016 monitoring period. * Mean value is indicative due to strong spatial variability and mixing effects.

Parameter	Reference Wells	Contaminated Wells
Well IDs	T26, V205, V206, V217, V219, V241	O6, O7, O8, T52, T54, T55, T57, T69, T70, T85, T86, T87, T89
Number of wells	6	13
Typical sampling depth (m b.g.l.)	8.6–11.6	10.8–11.7 (9 wells); 15.0–18.2 (T55, T57, T69, T70)
Apparent age (years)	26–43	1.3–14.1
Mean apparent age (years)	37 ± 4	~ 5 –10 *
Tritium (TU)	4.4 ± 3.2	>10 – $>10^3$ (measured), up to $\sim 10^5$ (historical peak)
R/Ra	2.8 ± 0.5	1.7–34.7
Hydrogeological setting	Upgradient, background conditions	Operational area, affected by releases
System type	Natural groundwater residence time	Anthropogenically influenced system

4. Discussion

4.1. Interpretation of Contaminated vs. Uncontaminated Wells

The potential sources of contamination are probably located in the vicinity of the Sanitary facility and laboratory west of the main building and on the route of the low-activity pipeline adjacent to Auxiliary building 1 (Figure 3). The east side of the main building does not contain any technological system that can function as a potential source.

The tritiogenic ^3He component derived from NPP-related tritium was corrected for the contribution of bomb-derived tritiogenic ^3He . Based on the reference wells and depth-adjusted estimates, the average bomb-derived contribution was 15.6 ± 13.6 TU, which was subtracted from the total tritiogenic ^3He in samples from the operating area.

Apparent ages in reference wells represent natural groundwater residence times, whereas in contaminated wells in the operating area they reflect transport times of anthropogenic (NPP-derived) tritium from the source area. The calculated ages are consistent with the timing of the known leakage events, indicating that the observed signals correspond to the migration of released tritium.

The wide range of R/R_a values (1.7–34.7) observed in the contaminated wells, compared to the relatively stable background ($\sim 2.8 \pm 0.5$), further supports the presence of a strong and spatially variable tritogenic ^3He source associated with NPP-derived tritium. Moreover, the variability observed within individual wells (e.g., O6: 1.3–6.0 years; T69: 10.8–11.7 years) demonstrates that vertical mixing and well-screen integration significantly influence the measured tracer signals, leading to composite apparent ages.

The observed spread in tritium concentrations and $^3\text{H}/^3\text{He}$ apparent ages is likely related to vertical mixing effects associated with long-screened wells. Depth-specific (packer-based) sampling demonstrated that bulk samples from long-screened wells integrate water from different depths, resulting in composite apparent ages influenced by both advective transport and vertical mixing.

This interpretation is further supported by lumped parameter modeling of the uncontaminated wells. Single-component models (e.g., piston-flow models implemented in TracerLPM) were able to adequately reproduce tracer concentrations in individual wells, but failed to consistently describe the aquifer system as a whole despite the close spatial proximity of the wells (Supplementary Figure S1). Acceptable fits across the dataset were only obtained using mixed lumped parameter models combining exponential and dispersion components (Lumpy), indicating substantial mixing and flow heterogeneity within the shallow aquifer. Model performance was generally high for ^3H , whereas lower goodness-of-fit values for ^3He likely reflect additional uncertainties associated with helium accumulation and subsurface transport processes.

4.2. Assumptions and Uncertainties of Age Interpretation

The following assumptions help explain variability in apparent ages between wells with similar construction parameters. The $^3\text{H}/^3\text{He}$ ages represent the time elapsed since groundwater recharge and are interpreted here as apparent tracer ages that may be affected by mixing processes.

The validity of the closed-system assumption is supported by regional recharge conditions. In central Hungary, recharge rates ($\sim 50 \text{ mm yr}^{-1}$ [8,42–44]) exceed the threshold typically required to prevent significant helium loss ($10\text{--}15 \text{ mm yr}^{-1}$ [5]), suggesting that the investigated shallow aquifer behaves effectively as a closed system with respect to helium.

However, the assumption of zero initial tritogenic helium for NPP-derived tritium introduces additional uncertainty. If tritiated water resides within closed technological systems prior to release, radiogenic ^3He may accumulate before entering the subsurface. The initial ^3He content at the time of release, therefore depends on the release pathway.

If the release occurs in the unsaturated zone, tritogenic helium may partially escape prior to recharge, resulting in negligible initial ^3He . In contrast, direct release into the saturated zone would prevent helium loss, leading to a non-zero initial ^3He concentration and thus a “virtual” initial age. Low uncertainties are derived from the high excess of tritogenic ^3He .

Consequently, the calculated $^3\text{H}/^3\text{He}$ ages presented in this study (Tables 2 and 3) should be interpreted as maximum apparent ages, particularly in cases where subsurface release pathways cannot be unambiguously constrained. The artificial elevation ($\sim 3 \text{ m}$) of the operating area results in effectively shallower sampling depths relative to the original ground surface, leading to younger sampled groundwater and reduced bomb-derived tritium contribution compared to the reference wells. As a result, the sampled groundwater near the power plant represents a shallower and therefore younger portion of the flow system. This implies that the tritogenic ^3He of bomb origin is also expected to be lower at these locations. This needs to be considered when comparing contaminated and reference wells.

4.3. Conceptual Picture

Anthropogenic tritium released from the nuclear facility acts as a transient tracer that allows direct observation of groundwater flow dynamics in the shallow aquifer. The results indicate that the natural flow system is overprinted by a younger and spatially heterogeneous tritium signal originating from leakage events. Apparent groundwater ages, therefore, represent mixed signals influenced by advective transport, dispersion, and vertical mixing processes.

These findings indicate that groundwater flow in the study area is not uniform but characterized by heterogeneous flow paths and depth-dependent transport processes, which are only partially resolved by conventional sampling. Overall, the results show that anthropogenic (NPP-derived) tritium can be effectively used not only as a contamination indicator but also as a tracer for constraining groundwater flow velocities, apparent ages, and mixing processes in shallow alluvial aquifers.

The interpretation of the calculated $^3\text{H}/^3\text{He}$ ages is subject to several limitations. The age calculation assumes negligible initial tritiogenic helium associated with the released tritium; therefore, any inherited ^3He component would lead to an overestimation of apparent ages. In addition, artificial ground elevation, engineered subsurface structures, and the strong hydraulic connection to the Danube may locally influence recharge conditions, groundwater flow paths, and mixing processes. Furthermore, the heterogeneous flow field and temporal variations in hydraulic gradients may result in mixing of groundwater with different travel times, causing deviations from the ideal piston-flow assumption and increasing the uncertainty of the calculated apparent ages. Consequently, the calculated values should be regarded as apparent ages reflecting tracer transport times rather than unique groundwater residence times. Nevertheless, the spatial distribution of apparent ages and their agreement with independently documented leakage events support the validity of the proposed interpretation.

5. Conclusions

This study presents the application of the $^3\text{H}/^3\text{He}$ dating method to investigate tritium contamination in a shallow alluvial aquifer at the Paks Nuclear Power Plant in Hungary, based on a four-year groundwater monitoring campaign (2013–2016).

The hydrogeological environment consists of a shallow unconfined aquifer composed mainly of sand and gravel deposits disturbed by artificial debris connected hydraulically to the Danube River. Groundwater samples were collected from monitoring wells and analyzed for tritium and noble gas concentrations.

The results show that uncontaminated reference wells represent natural groundwater residence times between 26 and 43 years (mean ~37 years), whereas wells within the operational area exhibit apparent ages between 1.3 and 14.1 years, reflecting the transport of tritium released during leakage events between 2005 and 2007.

The observed spatial variability in apparent ages highlights the influence of heterogeneous flow conditions, vertical mixing processes, and well construction. Depth-specific sampling using a packer system demonstrated that long-screened wells can significantly bias apparent age estimates by integrating groundwater of different ages. The derived ages should be interpreted as maximum apparent groundwater ages due to potential initial ^3He contributions and mixing effects.

This work demonstrates for the first time that anthropogenic tritium released from nuclear infrastructure can be quantitatively used to derive apparent groundwater ages using the $^3\text{H}/^3\text{He}$ method. These results highlight the potential of anthropogenic tracer signals to directly quantify groundwater flow dynamics and improve conceptual models of contaminant transport in hydraulically connected river–aquifer systems.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/hydrology13070174/s1>, Figure S1: Modeling of two lumped parameter models in parallel using the Parameter Steps function in Lumpy [24,39] Model A: Exponential Model (MRT 5 to 20, steps = 10). Model B: Dispersion Model (MRT 35 to 90, steps = 10 | Peclet Number 0.5 to 5, steps = 8). Mixture of A and B (0.1 to 0.9, steps = 16). The figures only displayed the results with the highest goodness of fit (Total Mix = 5.72, ^3H = 5.28 and $^3\text{He}_{\text{trit}}$ = 6.08).; Table S1: Summary of the modeled ages using a piston-flow model (PFM) using TracerLPM [41] compared to the apparent $^3\text{H}/^3\text{He}$ ages.

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Abbreviations

The following abbreviations are used in this manuscript:

PNPP	Paks Nuclear Power Plant
CWCH	Cold-Water Channel
PWR	Pressurized Water Reactors

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