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Infiltration of Lake Water into the Groundwater System Investigated by Tritium/Helium-3 Method: an Example from Wannsee and Lieper Bucht Area, Berlin, Germany

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Abstract: The T/^{3}He ages were calculated for groundwater samples collected from the locations Wannsee and Lieper Bucht area, southwestern Berlin, Germany. The calculated ages were used to study the infiltration of lake water into the groundwater system. The results showed that the distance of the wells from the bank of the lake does not affect the calculated ages, especially for the deep wells where all deep wells have nearly the same age. The shallow wells, which have ages nearly one year, have stable tritium of 11-12 TU, while the deep wells with ages nearly 24 years have stable tritium of 15-25 TU.

Keywords: T/3He age, groundwater system, Wannsee Lake, Lieper Bucht area

Introduction

Tritium and helium isotopes have been used as environmental tracers of groundwater flow in a variety of hydrogeologic settings. The ^{3}H/^{3}He ratio has been used successfully in dating groundwater less than 50 years old (Schlosser et al., 1988; Ekwurzel et al., 1994; Szabo et al., 1996; Plummer et al., 1998b).

In this study, ^{3}H/^{3}He dating method (Tolstikhin and Kamenski, 1969; Schlosser et al., 1988) was used to study the infiltration of lake water into the groundwater system. ^{3}H/^{3}He groundwater dating is based on the radioactive decay of tritium and the containment of the decay product ^{3}He in groundwater. ^{3}H/^{3}He directly yields a travel time and can be applied to a single sample (Schlosser et al., 1988). The groundwater ages were calculated from the ratio between tritiogenic helium and tritium as (Tolstikhin and Kamenski, 1969; Schlosser et al., 1988; Solomon et al., 1992; Aeschbach-Hertig et al., 1998) according to:

$$\tau = \frac{1}{\lambda} \ln\left[1 + \frac{^3He_{trit}}{^3H}\right] \qquad (1)$$

Where λ= 0.0556 yr^{-1} is the decay constant of ^{3}H and [^{3}H] the measured tritium concentration. It is usually expressed in tritium units (TU). 1 TU corresponds to a ^{3}H/^{1}H ratio of 10^{-18}. [^{3}He$_{trit}$] is defined as the fraction of the total ^{3}He produced by ^{3}H decay, it was calculated by the difference between the measured concentration [^{3}He$_{tot}$] and the concentrations of all other ^{3}He components [^{3}He$_{eq}$]: equilibrium, [^{3}He$_{exc}$]: excess air, [^{3}He$_{terr}$]: terrigenic):

$$^3He_{trit} = {}^3He_{tot} - {}^3He_{eq} - {}^3He_{exc} - {}^3He_{terr} \qquad (2)$$

The equilibrium concentration in water [^{3}He$_{eq}$] is determined by the water temperature and the ambient air pressure (altitude) during recharge. Solubility data for He and Ne isotopes are taken from Weiss (1970) and from Benson and Krause (1980). The atmospheric excess [^{3}He$_{exc}$] can be calculated from the Ne or also from the ^{4}He excess, provided the latter is exclusively of atmospheric origin. Isotopic and elemental compositions of the atmosphere are taken from Ozima and Podosek (1983). If the water sample contains ^{3}He$_{terr}$, which may be present in aquifers where the rocks are enriched in U or Th, or in groundwater samples in which young water has mixed with relatively old water containing terrigenic, and in some cases, mantle He. The analogous equations for ^{4}He and neon contain only the equilibrium and excess air terms as well as terrigenic ^{4}He. The equilibrium terms can be calculated from the mean annual ground temperature and the atmospheric pressure in the recharge area (Aeschbach-Hertig, 1998). Hence the neon balance equation can be solved for the excess air component. If the He/Ne ratio of the excess air is known, ^{4}He$_{exc}$ can be calculated and the ^{4}He balance equation can be solved for 4Heter. Finally, the ^{3}He/^{4}He ratios of

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the excess air and the terrigenic helium are needed to calculate the respective ^3He components and solve Equation 2 for $^3He_{trit}$ (Schlosser et al., 1989):

$$^3He_{trit}=^4He_{tot}.(R_{tot}-R_{terr})-^4He_{eq}.(R_{eq}-R_{terr})-(^4He/Ne)_{exc}.(Ne_{tot}-Ne_{eq}).(R_{exc}-R_{ter}) \quad (3)$$

Where R denotes $^3He/^4He$ ratios. The helium isotope ratio in solubility equilibrium (R_{eq}) is related to that in air (R_a) by $R_{eq}= \alpha R_a$, α is a fractionation factor of about 0.983 (Benson and Krause, 1980) and R_{terr} is the average $^3He/^4He$ production ratio in crustal rocks of 2×10^{-8} (Mamyrin and Tolstikhin, 1984).

The $^3He/^4He$ and $^4He/Ne$ ratios are usually assumed to be atmospheric; this assumption is not completely right, because of the measurements of noble gases in paleo groundwater shows that, the excess air can be fractionated relative to atmospheric air (Stute et al., 1995b).

The main objective of this study is to determine groundwater residence times by $^3H/^3He$ method in two locations, the location lake Wannsee and the location Lieper Bucht in Berlin, Germany and to differentiate between recently infiltrated lake water and older groundwater in the study area.

Hydrogeology of the field sites

The city of Berlin, Germany, is a densely populated area with 3.4 million inhabitants on 891 km². The city is located in the lowland region of a glacial valley affected by three major ice ages during the Pleistocene epoch, the Quaternary period, spanning the time between 1.8 million years ago and the beginning of the Holocene at 12.000 years ago. Due to the glacial erosion and drain of the ice melt, the rivers Dahme, Spree and Havel and their lake like widening Lake Tegel, Lake Wannsee and Lake Müggelsee formed this epoch. The three ice ages Elster, Saale and Weichsel left different sediments behind which are used as groundwater reservoirs today. These pleistocene sediments indicate a groundwater floor with an average thickness of ~150 m which can be divided into three aquifers:

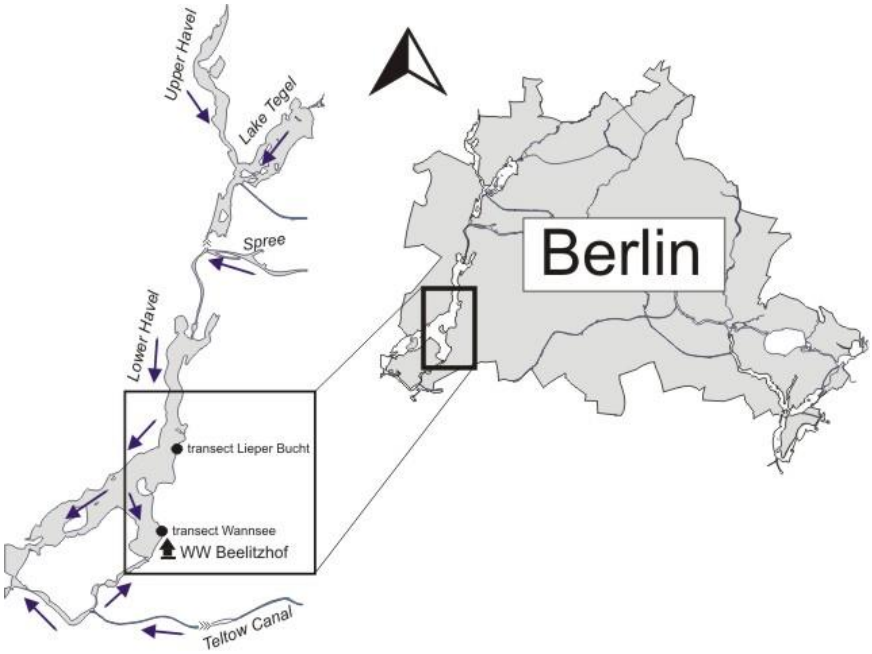

Figure 1: Wannsee and Lieper Bucht blocation

Aquifer 1, Weichsel ice age (70.000 years b.p.) with Holocene (12.000 years up to present): a sandy unconfined aquifer.
Aquifer 2, Saale ice age ((180.000 years b.p.) with Eem interglacial (100.000 years b.p.): fluvial and glacio-fluvial sands and gravels build an unconfined to confined aquifer
Aquifer 3, Elster ice age (480.000 years b.p.) with Holstein interglacial (320.000 a b.p.): fluvial sand and gravel in subglacial formed channels, silt, clay, lignite and mud, build a confined aquifer. These eroded channels locally cut a fourth aquifer with higher mineralized groundwater and even late Permian sediments. This fourth tertiary aquifer (Miocene to Upper Oligocene) with fine sand is not used for groundwater extracting, because this aquifer has low hydraulic conductivities and the local contact with late Permian (Zechstein) sediments leads to rise

salinity of groundwater and the water is no longer suitable for drinking water production.

Within the city area what is intensively used for different purposes, e.g., recreation, fishing, waterways and most important for gaining drinking water by lake bank filtration. In contrast to riverbank filtration, where the transit time of the water is in the range of days, in lake bank filtration the water needs several months to reach the production well. Since 1991 the drinking water is exclusively supplied by groundwater withdrawal supported by bank filtration. In order to replenish the groundwater reservoirs, Lake Bank filtration is enhanced and the amount of bank filtrates in drinking water is estimated to be 40-70 % nowadays.

Since 1991 the successive closures of lignite mines in the catchment's area of the river Spree resulted in a decreasing water level of the river Spree. The over-all flux is reduced from 45 m³/s in the years 1986-1995 to 28 m³/s in 1997. On the other hand, sewage treatment plants, which discharge a yearly total of 240 million m3 (7.6 m³/s) into the urban surface water system, represent important contributors for the water runoff. This high contribution has a considerable impact on the quality of the surface water.

Geology of Transect Wannsee

At the location transect Wannsee four aquifers build up the quaternary underground. The two aquifers from Weichsel ice age (aquifer 1) and Saale ice age (aquifer 2) cannot be defined and therefore seen as one consistent aquifer. These two aquifers have an average thickness of 20 to 40 meters and are separated by a ~ 10 m thick clay layer (Holstein interglacial) from the lower Elster-glacial aquifer. The Elster aquifer (aquifer 3) is almost continuous separated by an 8 m thick Elster-glacial silt layer building an upper and lower aquifer in between the third aquifer. The production wells of the waterworks Beelitzhof extract the water mainly from the Elster-glacial aquifer but some of the production wells (e.g. Br 4) have screens in different depths and pump additionally water from Weichsel- and Saale-glacial aquifer. The screens of the flat observation wells are in 5 to 7 m depth. The screens of the deep observation wells are in 60 to 80 m depth.

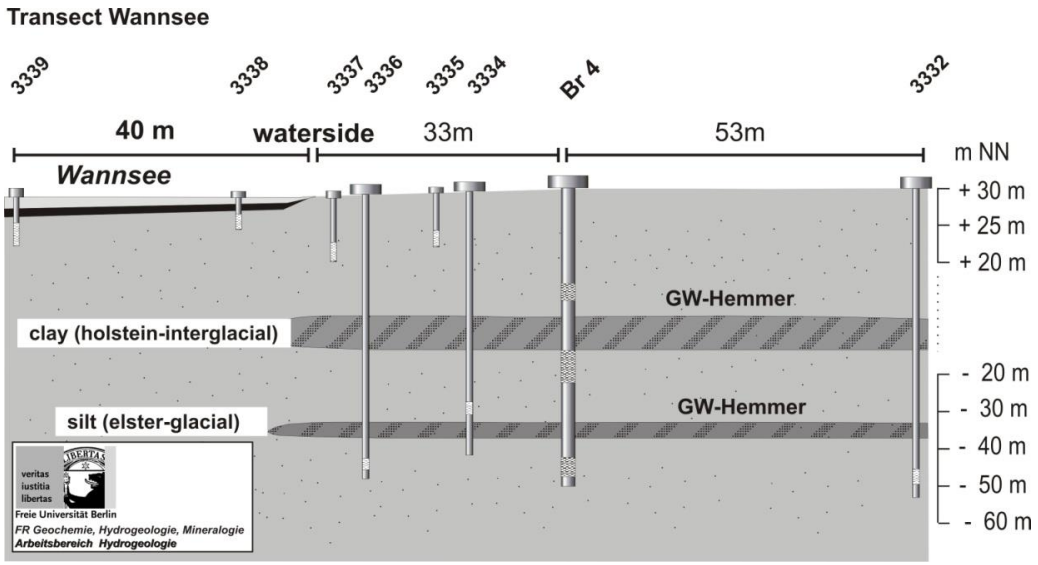

Figure 2: Cross section through the location transect Wannsee

Geology of Transect Lieper Bucht

At the location transect Lieper Bucht two aquifers build up the quaternary underground. The depth of the drill hole is 82 m. The upper aquifer has a thickness of 38 meters and consists of Holocene and Saale-glacial sands. A layer of 5 m thick Saale-glacial boulder clay interrupts this aquifer. A ~ 10 m thick layer of Holstein-interglacial clay separates the upper aquifer from the Elster-glacial aquifer. The Elster-glacial aquifer consists of Holstein-interglacial fine gravel in the upper part and of middle sands up to the depth of 82 m.

The screens of the flat observation wells are in 5 to 7 m depth. The screens of the deep observation wells and the production well (Br 14) are in 50 to 80 m depth.

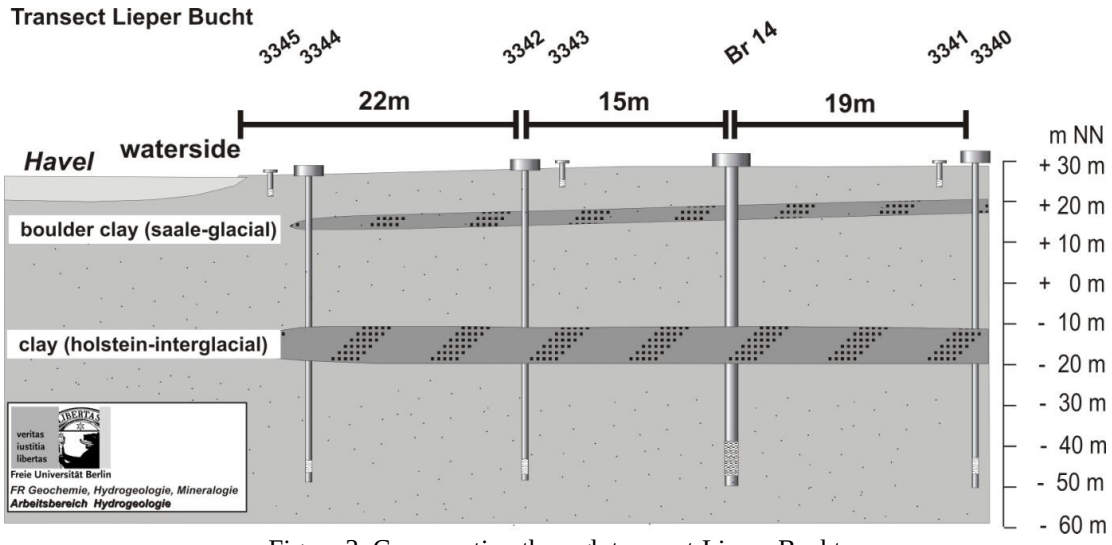

Figure 3: Cross section through transect Lieper Bucht

Sample collection and measurements

Seventeen samples of surface water and groundwater were collected for the analysis of helium and neon isotopes in the study area. The samples were collected in special pinch-off copper tubes (containing about 40 cc of water, and fitted with stainless steel pinch-off clamps at each end). The copper tube is fixed in an aluminum channel holding the stainless steel pinch-off clamps. By closing the clamps, the copper tube is shut absolutely gas tight. Clear plastic tubing was used for connecting the copper tube to the well or pump because one can visually observe whether air bubbles are present in the water line, all connections are airtight and will not come loose when back pressure is applied during closing of the copper tubes. Squeezing and bending the hoses or knocking against the tubing can remove the bubbles that tend to stick at connection. Sometimes, air is sucked in at connections of the tubing, such configurations must be avoided (always go from large to small diameter). Slight leaking of water out of the tubing may be tolerated. If bubbles form due to degassing of the water, an increase of pressure can often help to suppress degassing. In general, the pressure should be as high as the stability of the connections allows. The stability can be checked easily by putting a thumb on the outlet. Usually, the pressure is fine when the water jet after the copper tube reaches for several meters. Before sampling, the well should be sufficiently flushed (pump out at least once the volume of the borehole). During flushing, temperature and electrical conductivity may be checked to see if the values stabilize.

Water samples were analyzed for helium (He) and neon (Ne) isotopes at the noble gas laboratory of the Institute of Environmental Physics, University of Bremen. In the laboratory, the copper tubes were connected to a high vacuum system and permanent gases were separated from water and stored in glass ampoules. Leak checks and residual gas checks assured transfer efficiency greater than 99.9%. The glass ampoules were opened in a high vacuum inlet system. Water vapor transferred gases to a cryo system kept at 25 K to separate Ne from other gases. ^{20}Ne and ^{22}Ne were analyzed with a quadrupole mass spectrometer (Balzer QMG112A). Overall calibration with air aliquots and internal water standards assured accuracies for Ne concentrations in water higher than 1.0%. The 1r-standard deviation of ^{22}Ne/^{20}Ne was less than 0.2%. For most samples double measurements were conducted. More details on the measurement techniques can be found in Sültenfuß et al. (2004).

Tritium samples were collected in 1 litre glass bottles and were extracted from water samples by the gas extraction system described by Sültenfuß et al. (2004). The extraction efficiency of the system is at least 99.95 %. The degassed water was flame sealed in glass bulbs, which were stored in a freezer for nearly four weeks until a sufficient amount of tritiogenic ^{3}He had accumulated to allow determination of tritium content by the ^{3}He-ingrowth method. More details on the measurement techniques can be found in Sültenfuß et al. (2004)

Results and discussion

The analysis of ^{4}He and Ne concentrations

The observed ^{4}He and Ne concentrations (Table 1) appear to have two major sources: (1) gas exchange between lake water and the atmosphere, resulting in gas concentrations close to the solubility equilibrium with the atmosphere for the lake water temperature (2) complete dissolution of small air bubbles possibly caused by water table fluctuation in the recharge area (``excess air``, Heaton and Vogel, 1981). The plot of the concentration of helium versus Neon (Fig. 4) shows that some of samples have considerable

excesses of both noble gases relative to the solubility equilibrium with the atmosphere under condition typical for the aquifer. Some data point fall on or near the line that represents addition of excess air with an atmospheric Ne/He ratio such as TLB3343 (The data points from the transect Lieper Bucht are represented by the label (TLB) and the data points for the transect Wannsee are represented by the label (TW)). This strongly indicates that the excess air component is not fractionated (Aeschbach-Hertig, 1998). Several samples are marked by additional ^{4}He component of terrigenic origin such as wells TW Br4, TW3336 and TW3334 from transect Wannsee and TLBBr14 from transect Iieper Bucht. (Fig. 4 & Fig. 5).

For transect Lieper Bucht only the production well TLBBr14, which has a depth of 50-79 m has nearly 10% ^{4}He terrigenic. The wells (TLB 3340, TLB 3344 and TLB 3345) contain a very small portion of ^{4}He terrigenic comparing with the solubility equilibrium; the remaining wells have a zero ^{4}He terrigenic concentration (TLB 3341, TLB 3342 and TLB 3343) (Fig. 5).

For transect Wannsee, one can easily notice a distinct variation from a zero ^{4}He terrigenic concentrations in the shallow wells (TW3335, TW3337 and TW3339) to a very high ^{4}He terrigenic concentrations, which appear in the deep wells (TWBr4, Tw332, TW334 and TW336) (Fig.5), this variation in ^{4}He terrigenic can be explained as a result of different types of aquifers where the shallow aquifers belong to a Weichsel aquifer (sandy unconfined aquifer) and the deep aquifers belong to the Elster-glacial aquifer.

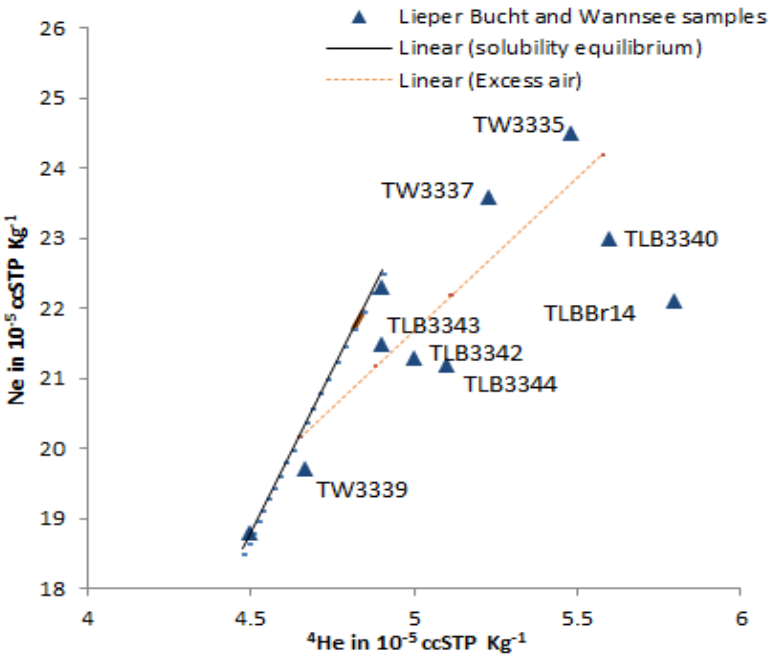

Figure 4. Neon concentration in 10^{-5} ccSTP/kg versus helium concentration in 10^{-5} ccSTP/kg.The solid line represents water in solubility equilibrium with atmosphere for a temperature range of 0°C to 20°C. The dashed line represents excess air at 10°C.

Test poin	Distance from lake [m]	Depth [m]	^{4}Hex10^{-5}[ccSTP/kg]	Nex10^{-5} [ccSTP/kg]	^{3}H [TU]	^{4}He$_{terr}$x10^{-5} [ccSTP/kg]	^{3}He$_{trit}$ [TU]	Age [years]	Stable T [TU]
TLB Br14	36	50-79	5.8	22.1	3.93±0.14	0.57±0.09	11.9±0.7	24.9±0.9	15.8
TLB 3340	55	76	5.6	23.0	ND	0.07±0.09	1.4±0.7	-	-
TLB 3341	46	6	4.9	22.3	10.15±0.2	0.0	0.3±0.3	0.5±0.4	10.4
TLB 3342	22	76	5.0	21.3	4.66±0.09	0.0	13.3±6	24.2±5.8	17.9
TLB 3343	24	12	4.9	21.5	10.70±0.20	0.0	0.4±0.2	0.66±0.4	11.1
TLB 3344	7	79	5.1	21.2	6.29±0.13	0.03±0.023	18.5±0.7	24.6±0.7	24.8
TLB 3345	3.5	3.6	4.5	18.8	11.51±0.23	0.13±0.07	0.6±0.5	0.9±0.7	12.11
TW Br4	33	80	22.1	24.1	2.4±0.1	16.26±0.23	8.0±1.6	26.24±2.8	10.4
TW 3332	70	76-80	9.9	24.8	ND	3.96±0.12	0.2±0.8	-	-
TW 3334	16	58-60	12.9	24.3	ND	7.07±0.14	1.2±1.0	-	-
TW 3335	17	6.2-8.2	5.5	24.5	11.1±0.2	0.0	0.5±0.4	0.79±0.6	11.6
TW 3336	10	73-75	34.7	25.3	ND	28.47±0.35	4.6±2.5	-	-
TW 3337	3	6.5-7.5	5.2	23.6	ND	0.0	ND	-	-
TW 3339	-40*	2.1-4.1	4.7	19.7	11.1±0.2	0.0	0.9±0.4	1.4±09	12

Table 1. Tritium, Helium, Neon Data, and calculated ^{3}H/^{3}He Ages for transect Wannsse and Lieper Bucht (ND means not detectable, * means the well lies inside the lake)

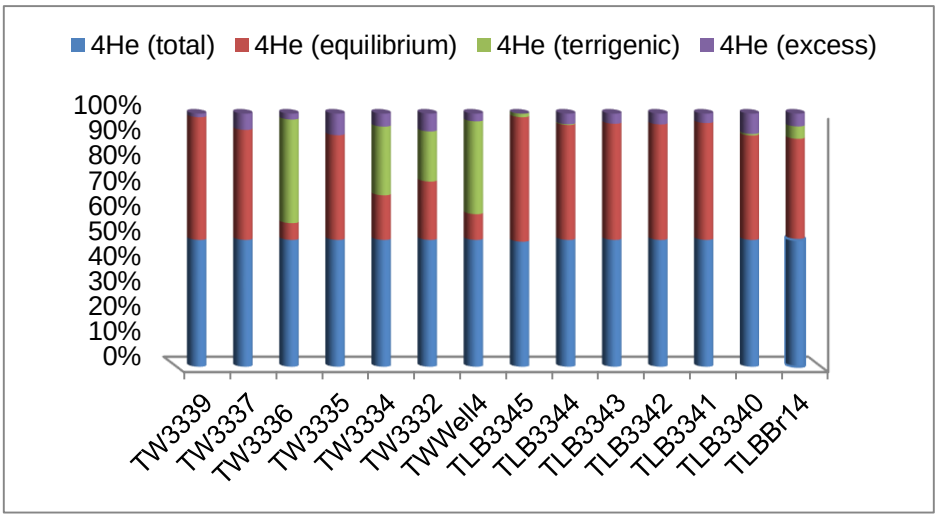

Figure 5. The analysis of 4He components for transect Wannsee and Lieper Bucht

In reality the zero ^{4}He terrigenic concentration means negative ^{4}He terrigenic concentrations, which have no meaning, this might be explained as follow: The assumption of excess air formed by complete dissolution of small air bubbles trapped in the soil during infiltration, and therefore the composition of excess air is identical to air, sometimes is not significant in most cases and the He/Ne ratio of excess air is not equal to the atmospheric value of 0.2882. Instead, it is lower, and to determine the appropriate value of He/Ne ratio additional information from other noble gases concentrations is needed. In the recent study only He and Ne data are available so that the assumption of zero concentration of ^{4}He terrigenic is considered to facilitate the calculation of ^{3}He tritiogenic by Eq. (3).

^{3}H/^{3}He ages for transect Lieper Bucht

The calculated ^{3}H/^{3}He ages did not show a distinct variation with the distance from the bank of the lake (figure 6), the distance of the wells from the bank of the lake does not affect the calculated ages, especially for the deep wells, which belong to the Elster-glacial aquifer where all deep wells have nearly the same age (~24 years).

All shallow wells, which belong to different aquifer (Holocene and Saale-glacial sands) have ages less than one year, the lowest age was observed in the well TLB3341, which is the furthest well from the bank of the lake, this is because the TLB3341well is inland of the production well, hence it does not lie along one flow path.

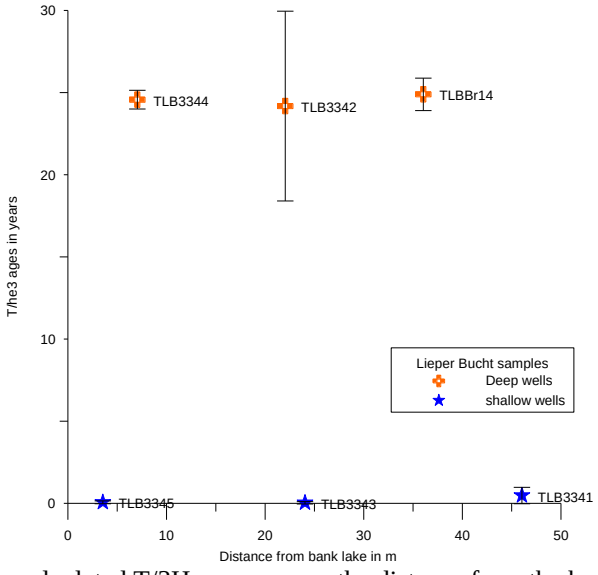

Figure 6: The calculated T/3He ages versus the distance from the bank of the lake

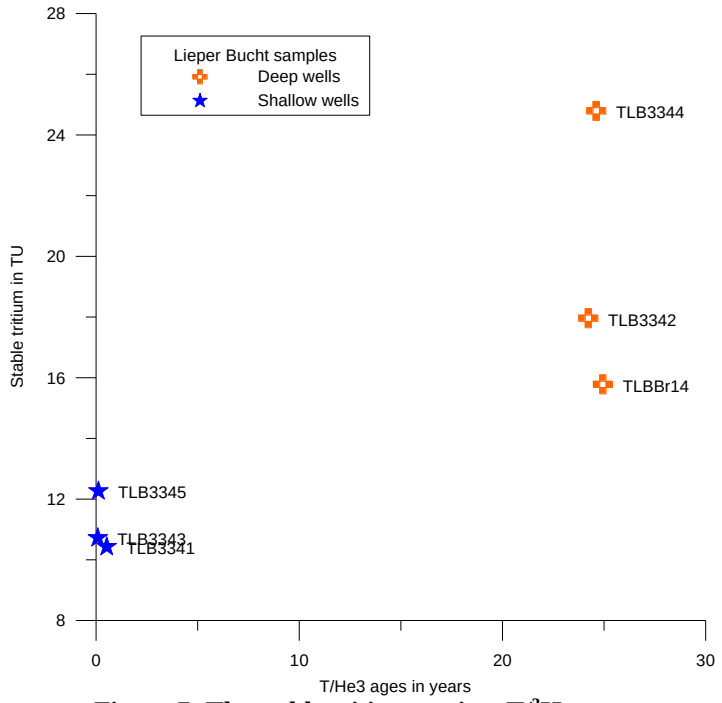

Figure 7: The stable tritium against T/^{3}He ages

The shallow wells, which have ages less than one year, have initial tritium 11-12 TU, the deep wells with ages nearly 24 years have initial tritium 15-25 TU (figure 7).

The main point of the calculated ^{3}H/^{3}He ages is that they definitely are the result of mixing of water from different sources other than bank filtration, which have different tritium and ^{3}He concentrations. The ^{3}H/^{3}He ages of a mixture of different water parcels is not a linear function of the ages of the individual parcels, especially if they have different tritium concentrations (Jenkins and Clarke, 1976). The addition of pre-bomb water virtually free of both tritium and tritiogenic ^{3}He would only dilute the concentrations of tritium and tritiogenic ^{3}He, but wouldn't alter their ratio. Therefore, the ^{3}H/^{3}He ages of the mixture would be the same as that of the young, tritium bearing component. In general the ^{3}H/^{3}He age of mixture deviates from the true mixing age towards the component with the higher tritium concentration (Aeshbach-Hertig et al, 1998).

Mixing has significantly influenced the ^{3}H/^{3}He ages from groundwater sample, which can be noticed by comparing the reconstructed original tritium content of the water samples (initial tritium) with historical records of the tritium concentration in precipitation, the tritium records from the IAEA (International Atomic Energy Agency) network are used to do this comparison. The data points from the sampling area are superimposed on the input function (Fig. 8), where the reconstructed tritium content of the water samples (stable or initial tritium) was compared with the historical records of tritium in precipitation, where one can recognize two different regimes: the first regime where the samples, which have ^{3}H/^{3}He ages less than one year fall on the input curve, this means that these samples were not influenced by mixing and affected by bank filtration, these samples (TLB 3341, TLB 3343 and TLB 3345) have initial tritium concentrations of 10-13 TU and the infiltration time is from 1999-2002. The second regime where the stable tritium fall below the input curves, these samples (TLBBr14, TLB3342 and TLB3344) have stable tritium of 15-25 TU, these samples were mixed by young water from another sources, the young water can leak down to reach the groundwater in the second aquifer through the windows found in the clay layers, which interrupt the two aquifers.

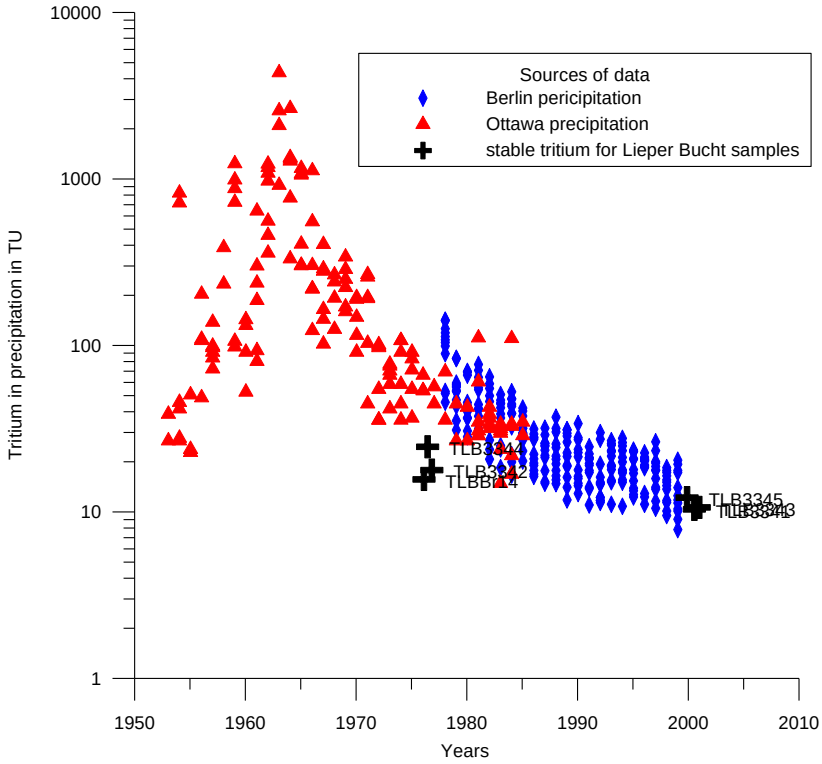

Figure 8: Comparison of the initial tritium concentration and the input function

$^3H/^3He$ ages for transect Wannsee

The $^3H/^3He$ ages were calculated in this part for only half of the samples because tritium is not measured for nearly 50% of the samples. The ages can be calculated for only three samples, the sample from the production well (TWBr4), which has an age (~26 years) and was mixed by young water from different sources where the production well has screens in different depths (figure 2), it has stable tritium (10.4 TU). The samples from shallow wells TW3335 and TW3339 have ages (~0.9 and ~ 1.5 year) and stable tritium (11.6 TU and 12 TU) respectively. One can notice that although the well TW3339 lies inside the lake, it has an age greater than the well TW3335, this can be explained by assuming that the well TW3335 was influenced by rain water and this is not possible for TW3339 because it is covered by a thick mud.

Conclusion

This study describes the time scales of groundwater systems in two locations; lake Wannsee and lake Lieper Bucht southwestern Berlin, Germany by using the $^3H/^3He$ method. This study demonstrates that $^3H/^3He$ dating method provides valuable information on the recharge dynamics and residence time of a lake bank infiltration on the groundwater system.

The calculated $^3H/^3He$ ages for water samples show distinct variation, where some samples have very young water less than one year, some others have water of ages about 25 years. These samples have a part which mixed with young water from different sources. The effect of mixing was discussed by comparing the reconstructed initial tritium ($^3H + ^3He$) content of samples with the temporal evolution of tritium in precipitation recorded in the study areas.

The occurrences of a high ^4He terrigenic in the groundwater samples gives a good indication to the presence of a significant component of old water e.g. sample TWBr4.

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