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**Summary of the Results and Interpretation of Tritium
And Noble Gas Measurements on Groundwater
Samples from the Perch Lake Basin Area**

**Résumé des résultats et interprétation des mesures du
tritium et des gaz rares sur des échantillons d'eau
souterraine de la zone du bassin de Perch Lake**

T. G. Kotzer

EACL

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RÉSUMÉ

On a prélevé, le long de la bordure centre-ouest du bassin inférieur de Perch Lake, un nombre restreint d'échantillons d'eaux souterraines des piézomètres à des profondeurs entre 8 et 17 mètres et à des distances entre 100 et 900 mètres vers l'aval de leur zone d'alimentation à proximité de la zone A. Les teneurs en tritium de ces eaux souterraines variaient approximativement de 100 à 2 800 UT. Les mesures des gaz dissous dans ces eaux souterraines indiquent des teneurs en ^4He et en néon qui s'approchent de celles des eaux souterraines récemment réalimentées; cependant, les teneurs en ^3He sont jusqu'à 100 fois supérieures, ce qui indique que ces eaux ont accumulé de l' ^3He tritiogène. On a calculé, grâce à la technique de datation $^3\text{H}/^3\text{He}$, des temps de séjour de l'eau souterraine de l'ordre de 29 ± 8 ans et des vitesses de l'ordre de 0,1 m/jour du réseau d'écoulement dans l'amas de sable central entre l'aire d'alimentation de la zone A et Perch Lake. Ces résultats, bien qu'ils soient fondés sur un très petit nombre d'analyses de l'eau souterraine, sont comparables aux estimations antérieures des temps de séjour et des vitesses de l'eau souterraine obtenues à l'aide de calculs de vitesse de Darcy, d'expériences de dilution dans les trous de sondage et de résultats d'essais par traceurs provenant d'études hydrologiques antérieures effectuées dans cette zone.

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ABSTRACT

Along the west-central margin of the Lower Perch Lake Basin, a limited number of groundwaters have been sampled from piezometers at depths of between 8 and 17 m and distances of between 100 and 900 m downgradient from their recharge location near Area A. Concentrations of tritium in these groundwaters varied between approximately 100 and 2800 TU. Measurements of dissolved gases in these groundwaters indicate concentrations of ^4He and neon approximating those in recently recharged groundwaters; however, the concentrations of ^3He are as much as 100 times higher, indicating the waters have accumulated tritiogenic ^3He . Using the $^3\text{H}/^3\text{He}$ dating technique, groundwater residence times on the order of 29 ± 8 years and groundwater velocities on the order of 0.1 m/day have been calculated for the flow system in the middle sand unit between Area A recharge and Perch Lake. These results, although based on a very small number of groundwater analyses, are comparable to earlier estimates of groundwater residence times and velocities obtained using Darcy calculations, borehole dilution experiments and tracer-test results from previous hydrogeologic studies in the area.

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1. INTRODUCTION

Tritium, occurring as HTO, in groundwaters can be used as a conservative tracer for hydrogeologic processes. In instances where the tritium input function into groundwaters recharging aquifers is accurately known, such as the large “spike” of anthropogenic tritium introduced into the hydrologic cycle in the 1950's and 1960's (Brown, 1989), measurement of the concentrations of tritium in an advecting groundwater can be used to calculate the time elapsed since recharge of the groundwater below the unsaturated zone, or the residence time of the groundwater (Solomon and Sudicky, 1991). In many cases, due to horizontal and vertical dispersion of dissolved groundwater constituents, this is not possible. In recent years it has been demonstrated that combined measurements of the concentrations of tritium ($t_{1/2} = 12.43$ years) and its decay product ^3He (tritogenic ^3He), along with other noble gases (^4He , neon) in groundwaters in relatively confined aquifers can yield reliable estimates of the residence times for a groundwater (Solomon et al., 1992; Solomon et al., 1993; Gascoyne and Kotzer, 1995 and references therein). The $^3\text{H}/^3\text{He}$ residence time calculated is independent of the initial tritium concentration in the water sample and, as such, is an apparent groundwater age. The $^3\text{H}/^3\text{He}$ residence time calculated can only be considered a true age if: i) non-tritogenic ^3He can be adequately corrected for, and ii) the groundwater sample is not a product of mixing of waters with different tritium and ^3He concentrations, or this process can be quantified. In a non-dispersive system where tritogenic ^3He is being quantitatively retained in the water mass, the $^3\text{H}/^3\text{He}$ residence time will agree with the true groundwater travel time, which is equivalent to the time of groundwater travel between recharge and the sampling point.

The technique for the sampling and measurement of tritium and noble gases (^3He , ^4He , ^{20}Ne , ^{21}Ne , ^{22}Ne) in groundwaters has been developed and utilized at Chalk River Laboratories and has yielded valuable information for several groundwater studies (Noack, 1995; Lee et al., 1995; Klukas et al., 1998a). Tritium and dissolved gas measurements were recently done on a limited number of tritium-enriched groundwater samples collected from the middle sand unit, downgradient along a hydrologic flowpath between Area A to Perch Lake at depths of between 8 and 17 m. The results of these isotopic measurements have been used to calculate residence times and groundwater velocities on the groundwaters using the $^3\text{H}/^3\text{He}$ dating technique and supplement residence times and groundwater velocities from previous tracer tests (i.e. Killey et. al., 1995).

2. RESULTS AND OBSERVATIONS

a) Tritium and Dissolved Gas Data

In June and October of 1998, samples of groundwaters were collected in the Lower Perch Lake Basin, from a number of piezometers which were screened off at various depths in the middle sand unit and located at varying distances downgradient from their point of recharge at Area A (Table 1- Figure 1).

Table 1
Hydrologic Information and Tritium Concentrations on Groundwaters
in Piezometers Sampled for this Study

Piezometer Number	Depth to Bottom of Piezometer (m)	Water Table	Tritium Concentration (TU)
QA3	12	surface	224
QA4	18	surface	11388
RB3	17	surface	2619
RB4	22	surface	4277
Q2	8	1 m below surface	347
Q3	12.5	1 m below surface	26055
PLS21	8	surface	152

In 1998 October, a subset of these piezometers were sampled and analyzed for their tritium and noble gas isotopic (helium and neon) concentrations (Table 2 - Figure 1). The piezometers were pumped to remove at least one standing volume of water from the entire casing to ensure fresh water for sampling. Tritium concentrations were measured using direct liquid scintillation counting with a Packard 1550 counter and 10 ml of waters mixed in a 1:1 ratio with scintillator. Dissolved gases in the groundwaters were sampled in copper tubes with He leak-tight pinch clamps and He and Ne concentrations and isotope ratios were measured using a MAPL 215-50 noble gas mass spectrometer. Details of these measurement procedures are outlined in Noack (1995) and Gascoyne and Kotzer (1995).

The concentrations of dissolved ^4He and Ne and isotope ratios of Ne in the groundwaters are similar to the theoretical solubility of He and Ne in a recharging groundwater which is in equilibrium with atmosphere at a temperature of 6°C (Table 2; Weiss, 1971). This suggests that the groundwaters are derived from recent recharge of surficial precipitation and also that dissolved gases in these samples were not fractionated during sampling. However, the isotope ratios of $^3\text{He}/^4\text{He}$ in the groundwaters are an order of magnitude higher than for normal groundwaters which have only equilibrated with atmosphere. Additionally, the ^3He concentrations in these groundwaters are significantly enriched ($\sim 100 \times$) relative to a groundwater at atmospheric equilibrium (Table 2). Although, ^3He can also be generated in the subsurface as a result of (n,α) reactions on dissolved lithium (i.e. $^6\text{Li} (n,\alpha) ^3\text{H} - ^3\text{He}$), the amounts of ^3He produced by such reactions are orders of magnitude less than the observed increases in ^3He in Perch Lake groundwaters (Gascoyne and Kotzer, 1995). Thus, the substantial increases in the concentrations of ^3He in the groundwaters are most likely from the decay of tritium to ^3He in the groundwaters over time.

Table 2
Piezometer Numbers and Concentrations of Tritium and Dissolved Helium
and Neon on Shallow Groundwaters from Lower Perch Lake Basin Area

Piezo#	Tritium (1)	³ He (2)	⁴ He (2)	³ He/ ⁴ He	Ne(2)	²⁰ Ne/ ²² Ne
QA3	100±23	3.2x10 ⁻¹²	8.1x10 ⁻⁸	3.9x10 ⁻⁵	3.1x10 ⁻⁷	
RB3	2869±140	1.3x10 ⁻¹¹	5.9x10 ⁻⁸	1.4x10 ⁻⁵	3.0x10 ⁻⁷	9.83
PLS21	91±23	1.0x10 ⁻¹²	7.4x10 ⁻⁸	1.6x10 ⁻⁵	2.5x10 ⁻⁷	9.89
6°C groundwater (3)		6.5x10 ⁻¹⁴	4.7x10 ⁻⁸	1.4x10 ⁻⁶	2.1x10 ⁻⁷	9.82

Remarks

- 1) Units are in tritium units (TU) where 1 TU = 0.1184 Bq/L
- 2) Units are in cm³ He or Ne/ mL water
- 3) Calculated concentrations of He and Ne in recharging groundwater at a temperature of 6°C (data from Weiss, 1971)

b) Calculation of ³H/³He Residence Times on Groundwaters

On the basis of the measured tritium and ³He concentrations for the Lower Perch Lake Basin groundwaters (Table 2), ³H/³He residence times have been calculated (Table 3; Figure 2a) using the equation:

$${}^3\text{H}/{}^3\text{He}_{\text{residence time}} = 1/l \times \ln (1 + ([{}^3\text{He}^*] / [{}^3\text{H}])), \quad (1)$$

where [³He*] = concentration of tritogenic ³He corrected for atmospheric equilibrium (6.5x10⁻¹⁴ cm³ ³He/ mL water); [³H] = concentration of tritium in the groundwater; $l = \ln 2 / 12.43$ (decay constant). In this calculation, it is assumed that the only source for the excess ³He is decay of tritium in the groundwaters and that the ³He has been quantitatively confined within the aquifer. This is generally satisfied when there is at least 1 m of water overlying the sampling depth and when the vertical rate of migration of the recharging groundwater is on the order of 2 cm/yr or better (Solomon et al., 1992, 1993).

From the results of these calculations (Table 3), it is apparent that, for two of the three groundwaters (RB3, PLS21), there is a general concordancy between residence times for Perch Lake Basin groundwaters derived using a variety of techniques. In particular, the overall ³H/³He residence time for groundwaters to transit from AREA A to Perch Lake calculated here (PLS21 - Table 3), agrees quite well with total residence time estimates of 22 years for groundwaters within the middle sand unit (Killey and King, 1997).

Table 3
Calculated Residence Times for Groundwaters from the
Perch Lake Basin Area using $^3\text{H}/^3\text{He}$ Technique

Piezometer	Tritium (TU)q	$^3\text{H}/^3\text{He}$ Residence Time (years; 1)	Residence Time Estimates from other Studies
QA3	100 ± 23	46.6±7	
RB3	2869 ± 140	18.8±5	10 (2)
PLS21	91 ± 23	29.4±8	22 (3)

Remarks

- 1) Errors calculated on the basis of precision of tritium analyses and using equation (1)
- 2) Residence time derived from flow velocities listed in Killey and King (1997; Figure 5)
- 3) Estimate from Killey and King (1997)

The much longer residence time for the groundwater sampled from QA3 (46 ± 7 years - Table 3) is puzzling in that it is the closest to recharge of the three piezometers investigated in this study and therefore should have the shortest residence time. On the basis of the very limited data set here, no definitive explanation can be offered for the apparent disequilibrium between tritium and tritiogenic ^3He in the waters. One possibility is that the screened interval for this piezometer is in a glacial till unit which has a much lower hydraulic conductivity than the sand units (Killey and King, 1997) and would therefore lead to a longer calculated residence time. Another possibility is that there has been lateral and vertical molecular diffusion of tritiogenic ^3He into the groundwaters in the vicinity of the piezometer at QA3 from the groundwaters having much higher concentrations of tritium and located closer to the center of the subsurface tritium plume. This possibility does exist since tritium concentrations in the groundwater from QA4, which is directly below QA3, are substantially higher (11388 TU; Table 1).

The effects of diffusion of tritiogenic ^3He from a tritium-rich water into the groundwaters sampled from QA3 have been calculated. Here, the tritium-enriched source for the excess tritiogenic ^3He in groundwaters from QA3 is assumed to have a tritium concentration of 10,000 TU which has decayed for 6 years, similar to the estimated residence time for groundwaters sampled from QA4 (Table 1). The diffusion coefficient for He in water ($3.16 \times 10^{-2} \text{ m}^2/\text{a}$ - Youngman, 1989) was corrected for flow in the saturated aquifer to give an apparent diffusion coefficient (D^* - Freeze and Cherry, 1979) of $1.58 \times 10^{-2} \text{ m}^2/\text{a}$ which was used in the equation:

$$\frac{C}{C_0} = \text{erfc} \frac{x}{2\sqrt{Dt}} \quad (2)$$

where C/C_0 = Relative concentration of the diffusing species at source (C_0) and at some distance away (C); x = distance; D^* = apparent diffusion co-efficient for He in aquifer; t = time (Freeze and Cherry, 1979).

Although only treated semi-quantitatively here, the results of the calculation (Figure 2b) indicate that for diffusion of tritogenic ^3He from a tritium-rich source to have been the cause of the observed tritium - tritogenic ^3He disequilibrium in the groundwaters sampled from QA3, the tritogenic ^3He source must be within 1 m of the sampling point or must have significantly higher tritium concentrations if it is located further away. The calculation shows that some diffusion of tritogenic ^3He is possible over a relatively short period of time, which could cause a disequilibrium between tritium and tritogenic ^3He in the groundwater samples.

Although, diffusion of He along concentration gradients in groundwaters has been discussed at length in the literature (i.e. Torgersen and Clark, 1985), its occurrence has generally been specific to subsurface diffusion of radiogenic ^4He , generated by α -decay of U and Th, and not tritogenic ^3He derived from decay of tritium. However, in many of these studies, groundwater tritium concentrations were much lower (40 TU and less). Given the large range of concentrations of tritium in the Lower Perch Lake Basin groundwaters, relative to natural groundwaters, tritogenic ^3He diffusion is possible. That this phenomena may be occurring in groundwaters sampled near the center of a groundwater tritium plume, is also evidenced by anomalously high concentrations of tritogenic ^3He which have been observed on groundwaters having low tritium concentrations and occurring immediately below the tritium-enriched contaminant plume at Area C (Kotzer and Mukai - unpublished data).

c) Groundwater Velocities

Groundwater velocities (Gw_v) are calculated here as:

$$Gw_v = \Delta L / {}^3\text{H}/{}^3\text{He} \text{ residence time} \quad (3)$$

where ΔL = distance between recharge point and piezometer sampled.

Here, using a residence time of 29 years for the groundwater at PLS-21 and equation (3), which is located at a distance of approximately 800 m from Area A recharge site, results in a groundwater velocity of approximately 0.1 m/day (0.08 ± 0.03 m/day). This is similar to the flow velocities for Lower Perch Lake Basin groundwaters estimated using Darcy calculations and tracer tests (summarized in Killey and King, 1997; Klukas et al., 1998b).

3. CONCLUSIONS

On the basis of measurements of tritium, helium and neon on groundwaters sampled from three piezometers in the middle sand unit of the Lower Perch Lake Basin, a series of groundwater residence times and groundwater flow velocities have been calculated. The results derived from

this study are in good accordance with those derived from previous groundwater studies in the area.

4. REFERENCES

Brown, R.M. 1989. A review of tritium dispersal in the environment. In Tritium and Advanced Fuels in Fusion Reactors, (eds. G. Bonizzoni and E. Sindoni), Editrice Compositori, Bologna, Italy; 557 - 575.

Freeze, R.A. and Cherry, J.A. 1979. GROUNDWATER. Prentice-Hall, Inc., Englewood Cliffs, N.J.; 604 pages.

Gascoyne, M. and Kotzer, T.G. 1995. Isotopic methods in hydrogeology and their application to the Underground Research Laboratory, Manitoba. AECL-11370; 101 pages.

Jackson, R.E., Johnston, L.M. and Inch, K.J. 1979. Geochemistry of groundwater and migration of contaminants in the Lower Perch Lake Basin. In, (eds. P.J. Barry) Hydrological and Geochemical Studies in the Perch Lake Basin: A Second Report of Progress, AECL- 6404; 169-177.

Killey, R.W.D and King, K.J. 1997. Update on the location of the leading edge of the Area A contaminant plume - 1997 March. RC-1959; 56 pages.

Killey, R.W.D., Klukas, M., Sakamoto, Y., Munch, J.H., Young, J.L., Welch, S.J., Risto, B.A., Eyvindson, S. and Moltyaner, G.L. 1995. The glass block experiment: Radionuclide release and transport during the past thirty years. RC-1513.

Klukas, M.H., Moltyaner, G.L., Takeda, S., Yamazaki, L. and Kotzer, T.G. 1998a. Advection-dispersion modeling of tritium and chloride migration in the Lake 233 drainage basin. RC-2132; 41 pages.

Klukas, M.H., Moltyaner, G.L. and Yamazaki, L. 1998b. Lower Perch Lake Basin three-dimensional groundwater flow model. RC-2014; 40 pages.

Lee, D., Kotzer, T.G. and King, K. 1995. Preliminary assessment of low- and intermediate level waste disposal in the Michigan Basin: Isotopic and Geochemical Measurements. COG-95-248; 51 pages.

Noack, 1995. Estimating groundwater velocity for a shallow unconfined aquifer using the $^3\text{H}/^3\text{He}$ dating technique: a comparison with other hydrogeologic methods. Unpublished M.Sc. Thesis, Trent University; 138 pages.

Solomon, D.K. and Sudicky, E.A. 1991. Tritium and helium-3 isotope ratios for direct estimation of spatial variations in groundwater recharge. *Water Resources Research*, v.27 (9); 2309 - 2319.

Solomon, D.K., Poreda, R., Schiff, S. And Cherry, J. 1992. Tritium and Helium-3 as groundwater age tracers in the Borden aquifer. *Water Resources Research*, v.28; 741-755.

Solomon, D.K., Schiff, S., Poreda, D. and Clarke, W.B. 1993. A validation of the $^3\text{H}/^3\text{He}$ method for determining groundwater recharge. *Water Resources Research*, v.29; 2951-2962.

Torgersen, T. and Clarke, W.B. 1985. Helium accumulation in groundwater: An evaluation of sources and the continental flux of crustal ^4He in the Great Artesian Basin, Australia. *Geochim. Cosmochim. Acta.*, v.49: 1211-1218.

Weiss, R.F. 1971. The solubility of helium and neon in water and seawater. *Journal Chemical Engineering Data*, v.16: 235-241.

Youngman, M.J. 1989. Dissolved gases and radioelements in groundwaters. Unpublished Ph.D. Thesis, University of Bath; 422 pages.

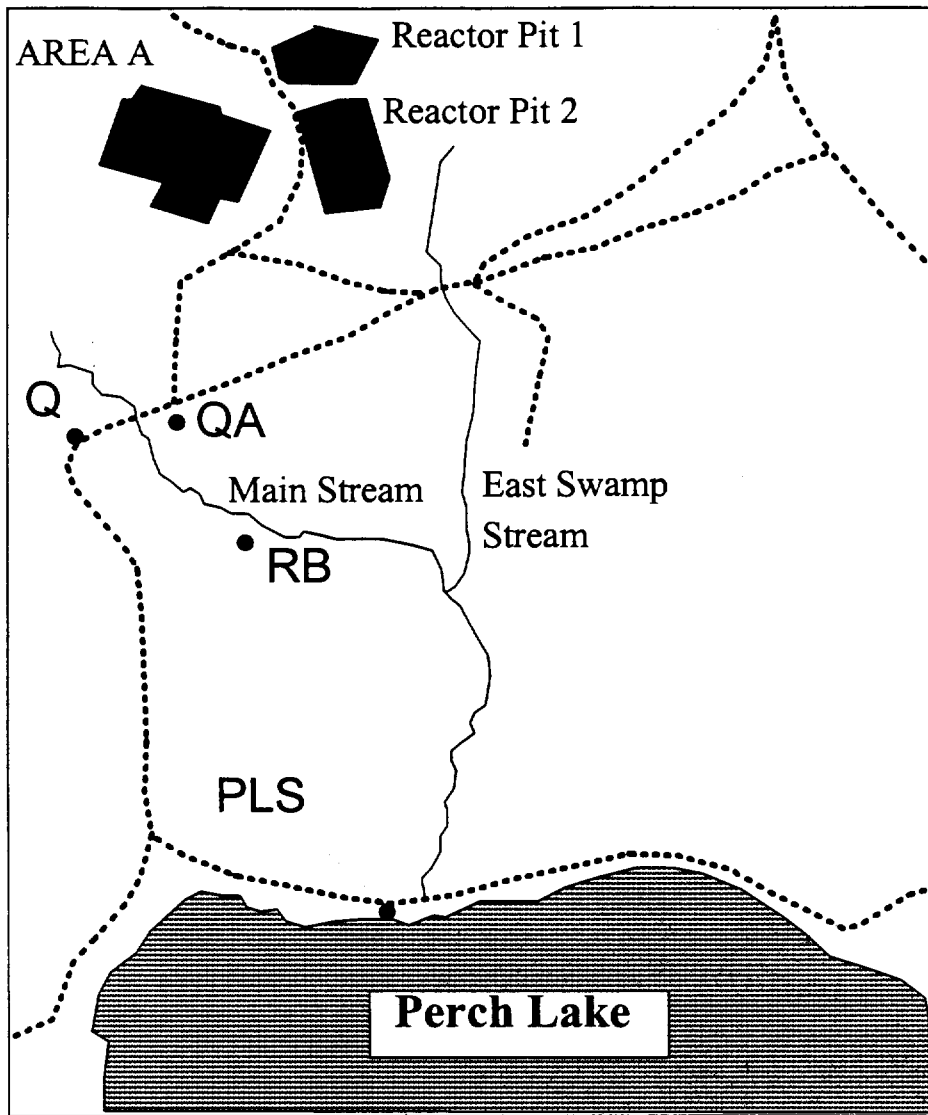


Figure 1: Map of the Lower Perch Lake Basin area showing the locations of piezometers where groundwaters were sampled for tritium and dissolved gas analysis. Figure adapted from Jackson et al. (1979).

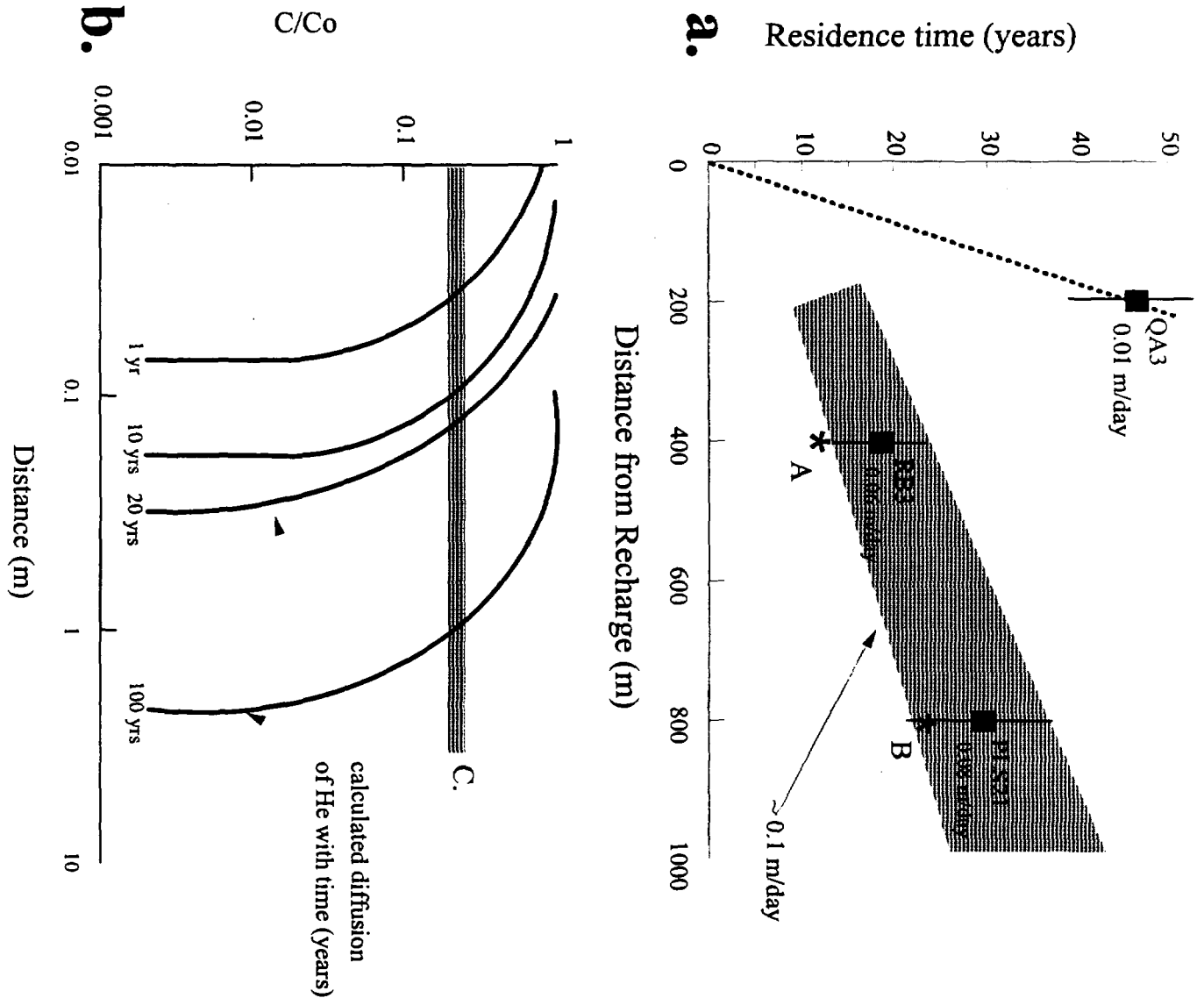


Figure 2: **a)** Comparison of calculated residence times and distance from recharge from groundwaters from this study. Data from Tables 2 and 3. Also shown are the groundwater residence times for waters in the middle sand aquifer at approximately 400 m. downgradient (A - Killey and King, 1997) and for the total length of the aquifer (B - Killey and King, 1997). **b)** Comparison of diffusion distances over time (shown by dark curved lines) with relative difference in concentrations calculated for tritogenic ^3He in Perch Lake groundwaters. Stippled, horizontal line labelled C. represents the C/C_0 ratio for the assumed source of the tritogenic ^3He (a 10,000 TU groundwater decayed for 6 years) and concentrations of tritogenic ^3He measured in the groundwaters sampled from QA3. The results indicate that at times of between 1 and 10 years, the excess tritogenic ^3He would have diffused less than 1 m from the source (intersection of line C. with 1 and 10 year diffusion curves).

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