Aquatic–Terrestrial Partitioning of Deep Groundwater Discharge Using Measured Helium Fluxes

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Performance assessments of nuclear fuel disposal vaults deep in the geosphere have generally assumed that all deep groundwater enters a lake or river and that no wetland or terrestrial discharge occurs. Animal licks in Ontario and Alberta, Canada, indicate that saline waters typical of deep groundwater do emerge in the biosphere. Helium gas, produced in situ in granitic rock, is used in this study to partition groundwater discharge. He flux comparisons were made on a flood plain with anomalously high He concentrations in the surface water of a creek and in the soil gas of an adjacent deer lick. This unique study, comparing direct flux-chamber methods with indirect methods using ping-pong balls as He gas collectors, indicates that 92% of the He produced in situ in the granite below this flood plain enters the creek and 8% enters the wetland. This partitioning is sensitive to the size of the terrestrial area considered and could be as high as 83% aquatic and 17% terrestrial at this location. Although this represents only one site, it suggests biosphere assessment models could use an aquatic–terrestrial partition of up to 80/20 for deep groundwater discharge unless site-specific data are available.

Introduction

Environmental impact statements (EIS) are required for many new industrial, transportation, and urban developments that could possibly affect the environment. The proposed disposal concept for nuclear fuel waste, deep in a pluton of intrusive rock on the Canadian Shield, is an example of a development that requires a detailed EIS (*1*). In fact, the EIS for this proposal may be one of the largest, most complex environmental, social, and political issues that will be addressed in Canada in the next decade.

Many environmental assessments are carried out using computer simulations to predict consequences of various impacts on both the human and nonhuman biota. Models of the biosphere have been constructed for a multitude of purposes, such as reactor operations (*2*), wetland productivity and management (*3*), soil erosion from agricultural practices (*4*), watershed acidification (*5*), and even the effects of climate change through increased CO_2 (*6*). These models all operate on very different temporal and spatial scales. The biosphere model, BIOTRAC, developed for the assessment of nuclear fuel waste disposal in Canada (*7*) needs to make quantitative predictions of the impacts to 10 000 years (*8*). This is primarily because the waste will be placed deep underground (500–1000 m) in rock, and it will take centuries to millennia for contaminants to diffuse through the host rock toward the biosphere.

Engineered and natural barriers will impede the transport of radionuclides dissolved in groundwater. Entry to the biosphere will probably occur in topographically low areas. The topographic lows will likely be streams, lakes, or wetlands that receive discharge from deep groundwater (*9*, *7*).

Until recently (*10–13*), little work has been published on methods of detecting locations of deep groundwater discharge. There has indeed been considerable debate as to whether deep groundwater actually discharges to a truly terrestrial area. Moose licks found in northwestern Ontario are saline seeps from the Nipigon sandstone formation and probably do not have a deep origin (*14*). Semi-terrestrial or wetland areas with saline soils (*15–17*) and supporting halophytic plants may however indicate seepage from a deep saline environment. Simple biosphere models that have been created outside Canada for deep underground storage or disposal of nuclear waste have not partitioned discharge to both an aquatic and a terrestrial area simultaneously; they tend to simulate each pathway separately and compare the consequences (*18*). BIOTRAC includes the possibility of terrestrial and aquatic discharge and estimates that discharge of deep groundwater will occur at least 90% of the time to a lake bottom and up to 10% of the time to a terrestrial area (*7*). No firm data exist to justify this partitioning of deep discharge.

Background. Helium (He) and radon (Rn) are the main gases produced by radioactive decay of U- and Th-bearing minerals in igneous rock formations. Helium is stable and

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accumulates with time, whereas Rn has a 3.8-day half-life and will reach a constant concentration in a closed system containing parent ^{226}Ra (an intermediate decay product). Both gases tend to diffuse out of their host minerals and, because of their solubility under pressure, accumulate in groundwater. In plutonic rocks, groundwater flow occurs almost exclusively along fractures, and extremely high levels of He and Rn have been observed in groundwaters at depth, especially in U- and Th-rich plutonic rocks (*19, 20*). Depending on flow conditions, both gases will tend to diffuse upward to the atmosphere where the fractures subcrop at the surface. In a groundwater recharge situation, the upflow of gases is limited, but in a discharge environment, high-concentration flows of He and Rn enter the atmosphere or mix with soil or surface water gases if the fracture outcrop is covered by water, sediments, or overburden.

The presence of these gases in soil and shallow groundwater has been used to detect subsurface bedrock fractures and to determine local hydrogeological conditions (*21–25*). Extremely high He concentrations (up to eight times atmospheric) in soil gases at a site (deer lick, Figure 1) heavily used as a soil lick by deer and in the water of an adjacent creek (*12, 13, 10*) prompted further study. The objective of this work was to measure the He fluxes in the adjacent aquatic and terrestrial areas to determine the aquatic–terrestrial discharge partitioning. We compare and discuss terrestrial He flux values obtained using direct trapping of soil gases at the soil–atmosphere boundary and using He soil–gas profiles as well as aquatic He flux values from surface water concentrations. Year-to-year variations in the He flux in the terrestrial area and seasonal and spatial variations in the creek are reported. Comparisons are made between the direct trapping of soil gas and a method using ping-pong balls as samplers. Finally, an aquatic–terrestrial discharge partition fraction is recommended for future biosphere modeling.

Study Site Description. The study site (50°16' N, 95°52' W) is located 5 km east of Lac du Bonnet, Manitoba, Canada (Figure 1, inset a). The site is situated in a valley floor, which often floods during spring-melt and high rainfall conditions (Figure 1). Overburden deposits in the valley floor are generally thin (<5 m), but they obscure the underlying Lac du Bonnet granite batholith. Isolated outcrops of granite are found in the upland area to the east and 0.5 km to the north where Boggy Creek descends over a waterfall. Peat accounts for most of the overburden, but the peat is underlain by silty clay toward the bedrock contact (Figure 1, inset b). Detailed geophysical, hydrogeological, and overburden studies (*12, 26*), define the extent and characteristics of the anomaly site. Geochemical evidence (*26*), based on Cl and ^{36}Cl analyses, states with little doubt that the groundwater at the base of the terrestrial deposit is characteristic of groundwater found >200 m deep in this batholith.

The terrestrial area of the site is small, about 100 m × 250 m in extent, and is covered by tall marsh grasses on a hummocky flood plain that forms the east bank of Boggy Creek (Figure 1). This terrestrial area is frequented by wildlife that appear to be eating soil and roots, exposing a bare peat area. This bare peat area is known as the 'deer lick' [after the 'moose licks' of northwestern Ontario, Canada, and the 'natural game licks' of Alberta and British Columbia where the animals are consuming salt from groundwater springs to satisfy sodium dietary deficiencies (*14, 17, 27*)]. The center of the deer lick is generally very soft and wet, indicating that groundwater may be surfacing at this point.

The aquatic area of the site is a 565-m stretch of Boggy Creek (formerly Dead Creek) characteristically marked by two distinct kinks in the creek, which possibly mark rock outcrop features that control the flow direction of the creek. The study area of Boggy Creek has widths between 10 and 22 m and depths ranging from 1 to 3.5 m (Figure 2).

Materials and Methods

Terrestrial Soil Gas Sampling Methods. The preliminary soil gas sampling of the site in 1991 was performed using standard probe and gas pumping equipment described by Gascoyne and Wuschke (*25*). In 1992, the site remained close to saturation throughout the year, and this method could not be used. An alternative technique using ping-pong balls, first developed by Dyck and Da Silva (*28*) for He analysis of lake sediments in the laboratory and later used by Stephenson et al. (*10*) for in situ measurements, was used. In 1993, gas flux chambers were employed to obtain direct flux measurements over the area when it was dry and to verify the results of the ping-pong ball method.

(a) Flux Chamber Method. Soil gas from the terrestrial site was captured in flux chambers and analyzed, providing a direct measurement of the emanation of He. The flux chambers measure the actual He flux to the atmosphere, which is more direct than predicting flux based on He concentrations in depth profiles in the soil and assuming a diffusivity value. As soil gases emerge from the ground, they were collected in 1-L stainless steel chambers [similar to the design of Raich et al. (*29*) and Reinhart and Cooper (*30*)], fitted with a rubber septum to allow sampling of the chamber gas with a 10-mL syringe (Figure 2). The gas samples were taken at regular intervals after installation, and the measured He concentrations were plotted against time. The change in He concentrations or flux density from the soil surface can be calculated as

$$J_s = (dC/dt)V/A \quad (1)$$

where J_s is the flux density (L He/m^2 soil·a), dC is the change in He concentration (μL of He/L of air), dt is the time from installation (a), V is the volume of chamber gas space (L of air), and A is the area of soil surface covered by the chamber (m^2 of soil).

(b) Ping-Pong Ball Method. This method involves placing ping-pong balls at standard depths in soils or sediments for several days (*28*). Helium dissolved in soil pore water surrounding the ball equilibrates with the air inside the ball and attains a concentration representative of that of the saturated soil. The ball is then removed, immediately sealed in a water-filled container that is impervious to He, and transported to the laboratory for analysis (*12, 11*).

In this study, ping-pong balls were mounted in both 3.5 m (long) and 0.8 m (short) 5 × 10 cm wooden stakes. The balls were placed in holes bored through the stakes and retained by a nylon mesh stapled to the stake on each side of the hole (*12*). The stakes are pushed into the wet peat to maximize contact of the peat with the ball and left in place for at least 7 days.

Short stakes, with one ping-pong ball at 50-cm depth and one immediately beneath the soil surface, were installed on two transects intersecting at the center of the deer lick.

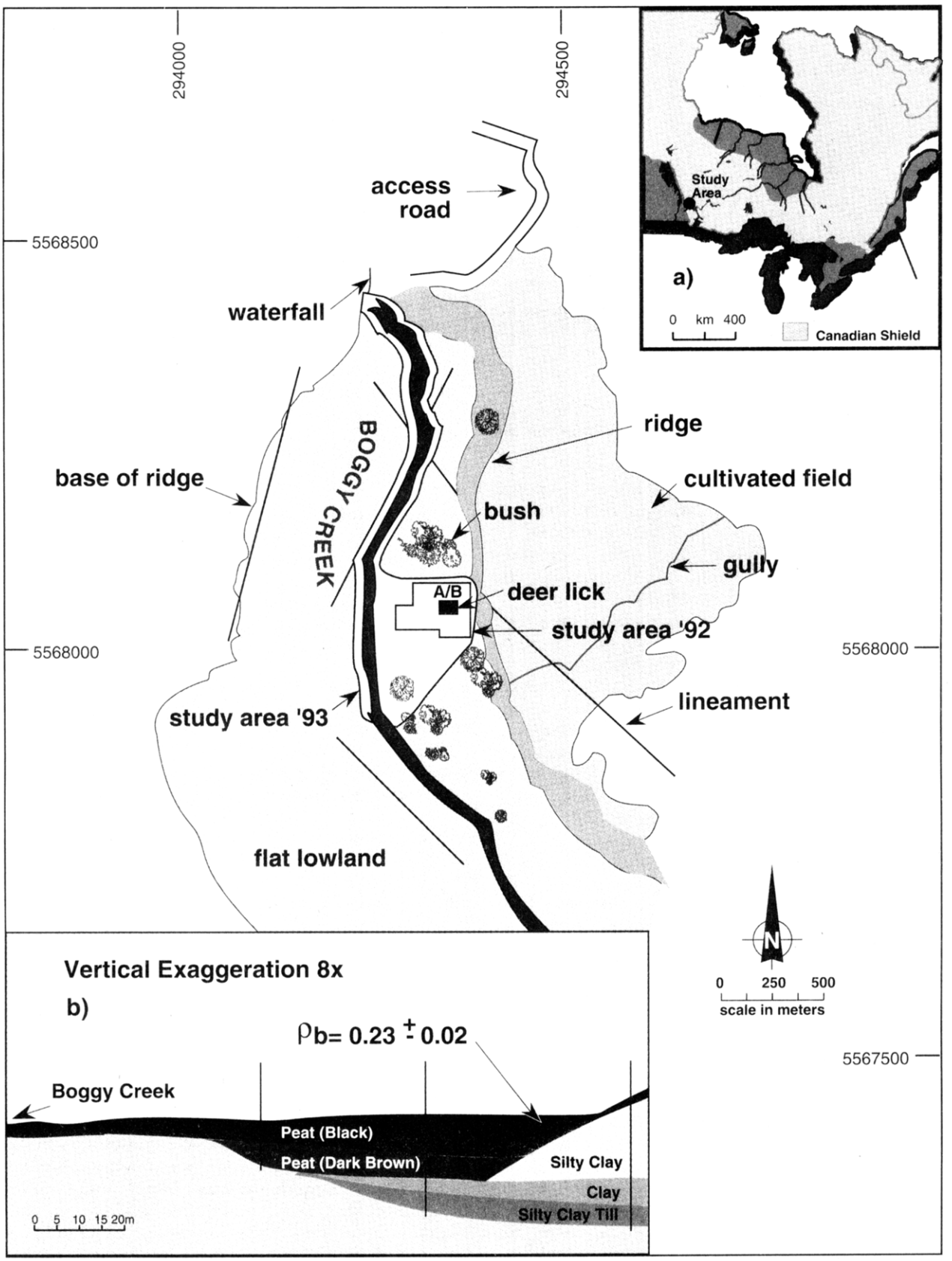

FIGURE 1. Study area location (a) in Canada with respect to the Precambrian Shield and (b) a vertical cross-section of the study area showing the overburden types and depths. Lineament lines (personal communication, Anton Brown) are identified by solid straight lines.

Each transect extended west to locations in the creek where high He was determined in preliminary studies (see Figure 3). The transects of short stakes at 10-m intervals were designed to give better definition of the He fluxes along the major regional lineament directions (NE–SW and NW–SE) and replicate values determined previously.

Two long stakes, with ping-pong balls located every 10 cm that had been driven to refusal (up to 3 m) in the previously mapped He hotspot (located at the intersection of the two transects) and that were left in place throughout

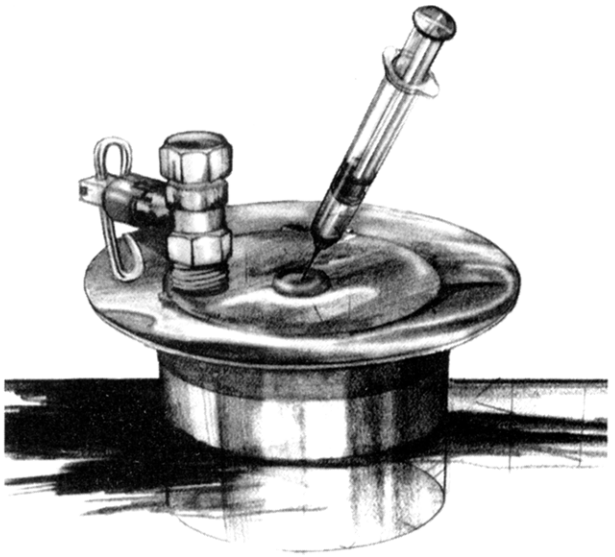

FIGURE 2. Schematic of the flux chamber used for collecting and sampling He gas.

the fall and winter of 1992 and into 1993 were also analyzed. The location of these long stakes remained the same; however, each time a pair of long stakes was removed for analysis, a second pair was installed. These long stakes provide He concentration profiles from the bedrock to the soil–atmosphere interface for several seasons.

Stakes were removed using levers and jacks as required. The ping-pong balls were removed and stored in steel-capped, water-filled glass containers (Mason jars) until analyzed (*12, 13*). The air in a ping-pong ball is a sample of the He soil gas dissolved in the pore water surrounding the ball. Helium concentration in the soil pore water, He_{diss} (μL of He/L of water), surrounding the ping-pong ball is determined as follows:

$$He_{diss} = 0.00899 He_{ball} \times 1000 \qquad (2)$$

where He_{ball} is the He concentration in the ball (μL of He/L of air).

The factor 0.00899 is derived from the ratio of the concentration of He in water, He_w, in equilibrium with air (0.047 μL of He/L of water at 10 °C) and He in air, He_a (5.24 μL of He/L of air). For an unsaturated environment, He concentration in the soil gas is the same as in the ball.

Aquatic Gas Sampling Methods. Water samples were taken at regular intervals along Boggy Creek both at the creek's surface and close to its bed. After the initial survey, sampling stations focused on the stretch of the creek, between stations 4 and 13b, which showed high He concentrations just NW and SW of the deer lick (Figure 3).

Collections were taken from a boat and by filling the sample bottles using a peristaltic pump and a plastic hose. A sampling stirrup was used to minimize sediment uptake (*10*) for the creek bed collections. The 773-mL glass sampling bottles were filled up slowly to prevent degassing, then 50 mL of water was withdrawn by syringe to create a known headspace, and the bottles were sealed with steel crown caps. The bottles were stored inverted to prevent air leakage before being analyzed in the laboratory.

Helium gas is sparingly soluble in water, so it preferentially partitions into air when a gas phase is present. The atmosphere (5.24 μL of He/L of air) equilibrates with surface waters at 10 °C to give concentrations of 0.047 μL of He/L of water. Excess He transported with groundwater into the waters and sampled into sealed bottles will equilibrate with the headspace gas, resulting in an increased He gas concentration in the headspace. By measuring the equilibrium He concentration in the headspace, it is possible to calculate the original aqueous He concentration in the water sample. Butt and Gole (*31*) provided an equation and the constants for this calculation:

$$He_w = (V_{hs}/V_w)[(He_{hs} - He_a) + (\beta He_{hs})] \qquad (3)$$

where He_w is the actual He concentration in water (μL of He/L of water), He_a is the concentration of He in air (5.24 μL of He/L of air), He_{hs} is the measured He concentration in headspace (μL of He/L of air), V_{hs} is the headspace gas volume (mL), V_w is the volume of water sample (mL), and β is the Bunsen solubility coefficient of He (0.00899).

The He concentrations in the surface water sample can be used to calculate an areal evasive flux, J_a, using the process of gas exchange at the air–water interface (*32*) and expressed as

$$J_a = k_{He}(He_{sfc} - He_{bkgd}) \qquad (4)$$

where k_{He} is the the gas exchange coefficient or piston velocity of He (m/a), He_{sfc} is the measured He concentration in the surface water (L of He/m^3 of water), and He_{bkgd} is the atmospheric equilibrium concentration of He (L of He/m^3 of air). The gas exchange coefficient, assuming the gas exchange process is controlled by the stagnant film thickness, z, is defined as

$$k_{He} = D_{He}/z \qquad (5)$$

where D_{He} is the the diffusion coefficient of He (7.24×10^{-2} m^2/a) and z is the surface water boundary film thickness (assumed to be 3×10^{-4} m).

Gas Sample Analysis. Helium concentrations of all gas samples were measured using a Veeco MS-18AB mass spectrometer (*12, 13*). Laboratory air (5.24 μL of He/L of air) and He standards in N_2 (11, 100, 1000 μL of He/L of N_2) were used to bracket 10-mL samples of soil gas (*12*). Chamber air samples were withdrawn for analysis using a 10-mL hypodermic syringe (Figure 2). For ping-pong balls and the headspace gas above the bottled water samples, air samples were withdrawn into syringes. For some of the samples, He concentrations were so large that the gas sample was first diluted (*13*). Two 10-mL gas samples were obtained per sample and analyzed together with standard gases in sequence (*12, 13*).

Results and Discussion

Terrestrial He Fluxes: Chamber Method. The He flux to the chambers was found to be sensitive to moisture conditions on this low-lying peaty surface. The He concentration in the chamber hovered close to atmospheric (5.24 μL of He/L of air) until the moisture content of the surface soil dropped below saturation, allowing the soil gases to emanate. The chambers were most effective for a short period in July when the site dried substantially. Preliminary tests indicated that at most locations installations of 2–3 days would be sufficient to measure above background He concentrations in the chambers accurately.

Twenty chambers, covering 0.08% of the deer lick area, monitored the He flux over two 4-day periods at the border and in the bare deer lick area (Figure 3) in the only two

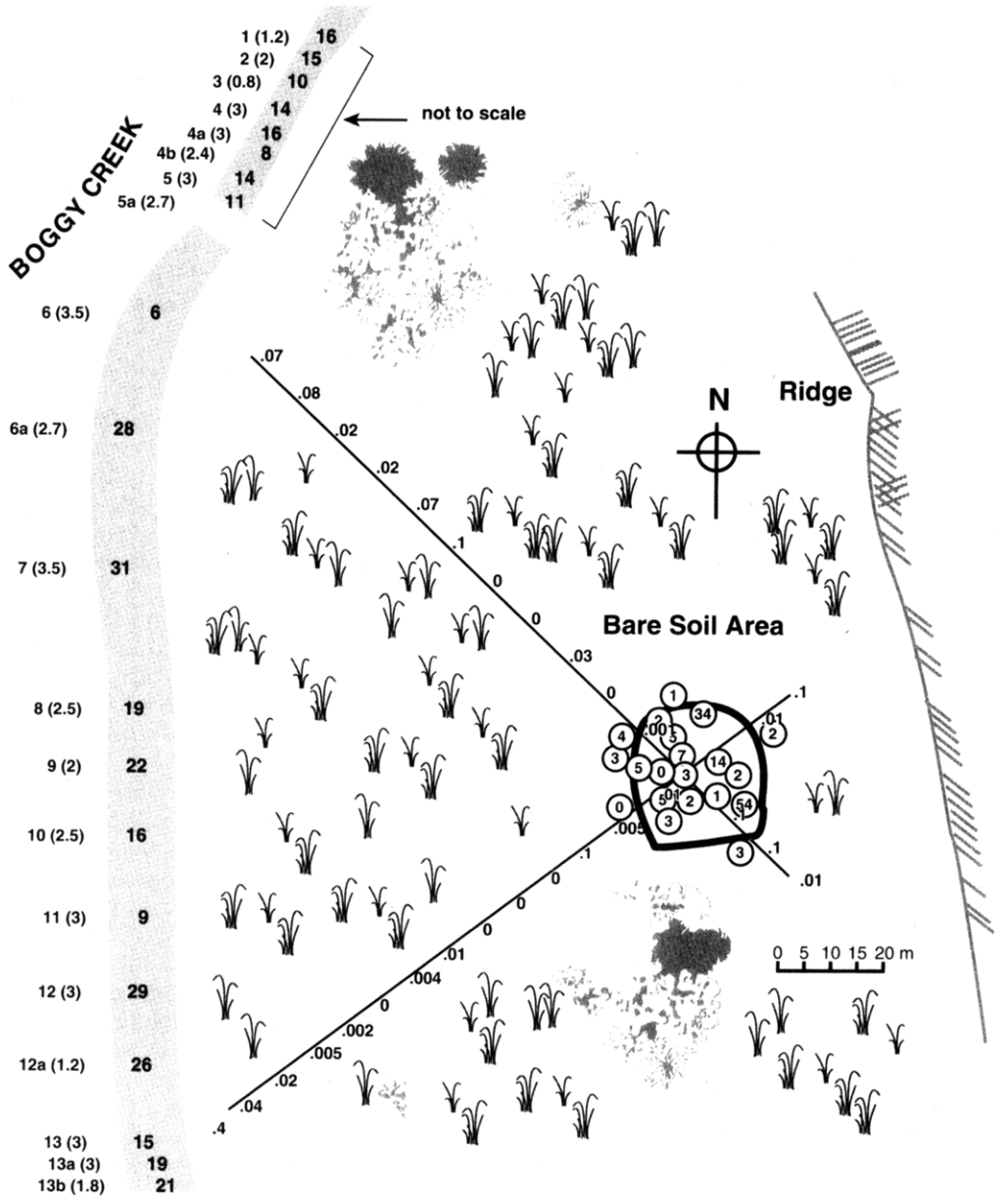

FIGURE 3. Enlargement of the study site showing the creek and the wetland, the location of the creek stations (their depths), and the terrestrial ping-pong ball stake transects. The heavy line demarcates the outer grassed area from the bare inner deer lick area. Open circles indicate the position of the He flux chambers, and the numbers inside are the He flux density values ($\times 10^{-3}$ L of He/m^2 of soil·a). The values every 10 m along the SW–NE and NW–SE transects are the flux density values (x 10^{-3} L of He/m^2 of soil·a) from the short ping-pong ball stakes, and the values on the creek are He flux densities ($\times 10^{-3}$ L of He/m^2 of water surface·a) from areal evasive flux calculations. All values shown are for July 20 and 21, 1993.

4-day periods without rain in July. The flux density values calculated using the chamber concentration data and eq 1 are shown in Figure 3, where dt is 350 000 s, the volume of the chamber V is 1 L, and the area of soil surface enclosed in the chamber is 0.0154 m^2. These flux densities vary from 0 to 54 $\times$ 10^{-3} L/m^2·a giving a maximum flux of 8.3 $\times$ 10^{-4} L/a in the SE corner of the lick (Figure 3). The average flux density from the entire deer lick was 9.75 $\times$ 10^{-3} $\pm$ 15.31 $\times$ 10^{-3} L/m^2·a, producing an average He flux of 3.5 L/a over this 359.4 m^2 area. The largest measured fluxes were not at the very center of the lick, but toward the edge of the bare area (Figure 3); the heavy line shows the devegetated central portion of the deer lick. The very high moisture content of the peat in the deer lick center explains the lower fluxes there.

This average flux in 1993 is almost 80 times smaller than the average flux of 270 L/a predicted from He concentrations using long ping-pong ball stakes inserted at the center of the deer lick in 1992, assuming a tortuosity factor of 0.5 and using the diffusion coefficient for He in air when the site was drier (*13*). Although the site appeared dry in 1992, this value may be an overestimate because we assumed all the pores to the depth of the stakes were air-filled. The diffusion coefficient of He in air is almost 30 000 times larger than the diffusion of He in water, and although the peat appeared to be cracked and dry, the true diffusion coefficient is probably between these two extremes.

Two conclusions can be drawn from these data. The use of chambers to determine the He flux at this site and other low-lying sites is difficult and heavily dependent upon

the moisture status of the soil. Based on the comparison with the previous flux data and the very wet nature of the site, these chamber measurements probably represent a minimal He flux at this wetland site. Similarly, because we used the diffusion coefficient of He in air to determine the flux previously, the previous flux estimates may be biased high. This would put the extremes of He flux at this terrestrial wetland site between 3.5 and 270 L/a. With a groundwater He concentration of 10 mL of He/L of water (*13*), this suggests the discharge of groundwater from this site is between 350 and 27 000 L/a.

Terrestrial He Fluxes: Ping-Pong Ball Method. The flux of He from the deer lick surface can be determined using Fick's first law, describing simple diffusion of gases in one direction under steady-state conditions and stated as

$$F = D(dC/dz) \quad (6)$$

where F is the He flux per unit area (m^2) per unit time (a), dC is the change in He concentration per unit length, dz (m) in the direction of diffusion, and D is the diffusion coefficient (m^2/a) for He through the matrix. The He concentrations in the peat pore water from the ping-pong balls at 0 and 50 cm provide the gradient of He described by the second term on the right-hand side of eq 6. Since there was usually standing water on the site, we assumed the movement of He through the surface peat was predominantly by diffusion through the pore water and used the molecular diffusion coefficient for He in water of 7.24×10^{-2} m^2/a (*33*). Comparison of a measured flux using a chamber with that calculated from this He gradient and diffusion coefficient at a stake position in close proximity indicated that the two methods gave different results. The flux calculated using Fick's first law was almost 10-fold smaller than the measured flux. This difference must lie in the assumed diffusion coefficient value. Flooding and large rainfall events could trap air in the peat deposit, and although it appears saturated, a small percentage of the pore space may be air-filled. The diffusion coefficient of He in air [2.05×10^3 m^2/a (*33*)] is 4 orders of magnitude larger than that in water (7.24×10^{-2} m^2/a). Using the flux measured in a chamber in close proximity (within 2 m) to a ping-pong ball stake and the gradient observed across the 50 cm of wet peat between the two ping-pong balls suggests an effective diffusion coefficient of 1.04 m^2/a. Particle (0.84 g/cm^3) and dry soil bulk (0.24 g/cm^3) densities determined for the top 20 cm of the lick peat were used to calculate a total pore space volume (0.72 cm^3 of pore/cm^3 of peat). This effective diffusion coefficient, obtained through calibration with a direct measurement, was combined with the pore space volume to determine what percentage of the pores was filled with air and what percentage was filled with water to achieve this diffusion coefficient. According to this calculation, the pores were 0.02% air-filled. This is reasonable because at the time of this measurement there was no standing water on the site and conditions were conducive to surface evaporation. This effective diffusion coefficient was used in eq 6 to calculate the fluxes for the remaining positions on the SW–NE and NW–SE transects that joined the bare deer lick area to the creek (Figure 3).

These short stake transects reveal that He seems to be diffusing to the surface at isolated locations rather than evenly along either of the two major lineament directions sampled. It is possible that our transects, even though they were guided by both electromagnetic and electrical conductivity surveys, missed the lineament transporting He and saline groundwater to the site. However, based on these data and previous mapping of the site under drier conditions, it seems more likely that pipe flow occurs in the near-surface rock and He only vents at selected locations along the feature that exists at depth. Perhaps the fracture infill material only allows passage of He and deep groundwater at discrete locations of infill weakness, and the deer lick is one of those isolated locations.

The average flux from the two long stake profiles in the summer of 1991, when the surface 1 m of soil was unsaturated, was 2.1 $L/m^2 \cdot a$ (*13*). This flux of 2.1 $L/m^2 \cdot a$ assumed that molecular diffusion of He in soil air was the dominant transport mechanism, there was no effect of tortuosity, and the pore space was 72%. Long stakes containing duplicate ping-pong balls at every 20 cm depth have been kept in place since 1991. On several occasions these stakes have been retrieved and the balls analyzed. Fluxes were calculated for the surface 50 cm of peat using duplicate long ping-pong ball stakes from August, October, and December 1992. Diffusion coefficients for He in air were used when this depth was unsaturated (August and December). The diffusion coefficient for He in water was used when it was saturated (October). We assumed there was no effect of tortuosity. The values were 205 and 277 $L/m^2 \cdot a$ (August,dry), 0.14 and 0.57 $L/m^2 \cdot a$ (October, wet), and 135 $L/m^2 \cdot a$ (December, dry), and 0.27 $L/m^2 \cdot a$ (December, wet). These values are not as divergent as they seem. A 1000-fold difference in flux is achieved when <4% of the porosity becomes air-filled, making gas fluxes difficult to predict in surfaces that dry and wet continuously.

The flux from the surface of the lick is a flux off a static surface and can be described by classical diffusion and stagnant boundary film theory when the deer lick is flooded. When the water table drops, the surface peat still contains a lot of water (>50% by volume). This water contains a lot of He, especially in the lick area; however, it is connected to a large network of open pores. This water and peat degas very rapidly, creating a large flux upon drying. Similarly, when the peat refloods, the surface layers are depleted in He, and a strong gradient is set up between the He-depleted surface peat and the basal peat bathed in He-rich groundwater. If the surface flux is measured over adequate lengths of time, inclusive of wetting and drying cycles, the surface flux becomes representative of the deeper basal flux of He to a terrestrial area.

Aquatic He Concentrations and Fluxes. Initially, 13 locations along the creek were sampled to determine surface and bottom water He. An additional seven stations, identified "a" and "b" following the station number (Figure 3), were added in the proximity of the stations with elevated He concentrations in the water and between the two terrestrial transects. Sampling three times at the center of the Creek was carried out coincident with sampling on the deer lick. Water samples were obtained on June 29, July 21, and August 10. The He concentrations in the surface and bottom water did not vary significantly over time; the results for July 21 are shown in Figure 4. The surface samples consistently showed He concentrations above those in equilibrium with the atmosphere (~0.074 μL of He/L of water). Stations 5, 6, 12, and 13 had high He concentrations in the bottom water. The elevated He concentrations found in this survey are at the same positions

TABLE 1

He Flux Density ($\times 10^{-3}$ L/m^2·a) and Flux (L/a) Values for Creek Station Segments for Three Sampling Times in 1993

		flux density			flux		
station	area	June 29	July 21	August 10	June 29	July 21	August 10
1	888	14.2	15.7		12.6	14.0	
2	1052	28.3	14.8		29.8	15.6	
3	764	18.6	10.4		14.2	7.9	
4	722	8.2	13.9	14.4	5.9	10.0	10.4
4a	450	11.3	15.4	15.1	5.1	6.9	6.8
4b	262	17.0	7.9	5.9	4.4	2.1	1.5
5	315	11.0	13.5	9.2	3.5	4.3	2.9
5a	336	21.0	10.7	4.1	7.1	3.6	1.4
6	379	85.0	5.7	12.1	32.2	2.1	4.6
6a	404	8.8	27.4	5.6	3.6	11.1	2.3
7	257	11.3	30.6	10.0	2.9	7.8	2.6
8	152	19.8	18.6	14.7	3.0	2.8	2.2
9	195	17.6	22.0		3.4	4.3	
10	231	18.3	16.4		4.2	3.8	
11	238	11.0	8.5	12.7	2.6	2.0	3.0
12	239	18.0	29.3	10.7	4.3	7.0	2.6
12a	182	19.8	25.8	17.4	3.6	4.7	3.2
13	264	18.0	15.4	7.7	4.7	4.1	2.0
13a	519	19.5	19.2	13.8	10.1	10.0	7.2
13b	616	21.4	20.5	16.6	13.2	12.6	10.2
total	8465	398.1	341.7	170.0	170.4	136.7	62.9
		19.9 ± 16.1	17.1 ± 7.1	11.3 ± 4.2	8.5 ± 8.5	6.8 ± 4.2	4.2 ± 3.0
		(n = 20)	(n = 20)	(n = 15)	(n = 20)	(n = 20)	(n = 15)

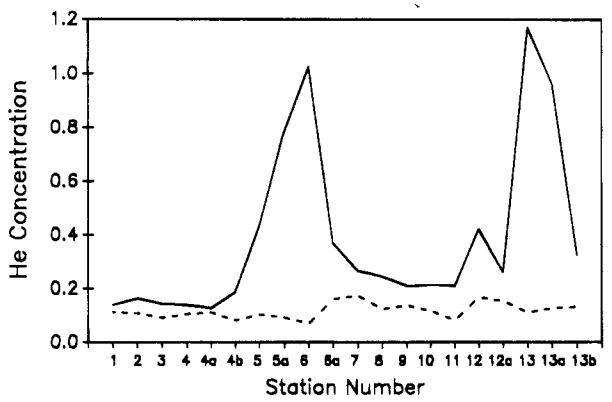

FIGURE 4. Boggy Creek surface (dashed line) and bottom water (solid line) He concentrations (μL of He/L of water) by station along the creek. The atmosphere (5.24 μL of He/L of air) equilibrates with surface waters at 10 °C to give concentrations of 0.047 μL of He/L of water. Analytical precision is typically within ±0.1 μL/L.

as previously reported by Stephenson et al. (*10*). The two positions occur where the creek diverts westward for a straight stretch and then corrects itself (Figure 1). Stephenson et al. (*10*) also showed high He concentrations correlated well with changes in the direction of creek flow. This could be associated with subsurface features or lineaments that have influenced the path of the water. Creek depths at stations 5, 6, 12, and 13 were 3, 3.5, 3, and 3 m, respectively, which were some of the deepest positions (Figure 3) and deeper than the mean (2.5 ± 0.75 m, n = 20).

The deer lick connects with a NW–SE line from station 6, the location where the highest flux was measured, and a SW–NE line from station 13 (Figure 3), following the direction of major subcrop fractures in the area (Figure 1). This is further speculation that these locations of elevated He, both aquatic and terrestrial, are related to deep groundwater exiting the surface through fractures below our study site. However, it is possible that these terrestrial and aquatic sites are not connected but are a result of different permeable zones of one or two fractures in the granite.

The areal evasive flux densities for each of the three sampling times for the creek stations are given in Table 1. Some of the highest flux density values were found between stations 5a and 13b, in the straight stretch of the creek, and were measured early in the season when drier conditions prevailed. On August 10, the creek was still flushing twice the normal amount of rainfall for July as well as abnormally high rainfall for early August. Average areal evasive flux density values for the 11 stations from 6a to 13b for June 29 and July 21 were $1.7 \times 10^{-2} \pm 4.2 \times 10^{-3}$ and $2.1 \times 10^{-2} \pm 6.6 \times 10^{-3}$ L/m^2·a, and the mean flux density value at nine stations on August 10 was $1.2 \times 10^{-2} \pm 4.0 \times 10^{-3}$ L/m^2·a. The highest flux measured in the creek was 8.5×10^{-2} at station 6 on June 29. Most stations only vary 3–5-fold during the season; however, station 6 varied 15-fold between the first and second sampling. If we join station 6, which is at a flow divergence in the creek, with the He hotspot on the deer lick, this line trends NW–SE. This is the direction that the geophysical data (*12*) indicated had lower total magnetic field strength, indicative of leaching of the bed rock, and this transect is aligned with lineaments of the area (Figure 1).

Aquatic/Terrestrial Discharge Partition. The total He flux from the creek using measured creek widths and lengths from a digitized air photo was 170 and 137 L/a on June 29 and July 21, 1993. The total flux from stations 4–13b on August 10, 1993, was 63 L/a (Table 1). The total flux for the bare deer lick area (359.5 m^2) on July 20, using the average of the 20 chamber values, was 3.5 L/a. The NW–SE transect connects station 6, where the largest He flux was observed on the creek, with the largest measured terrestrial He flux near the SE corner of the deer lick. The aquatic flux density at station 6 on July 21 was 5.7×10^{-3} L/m^2·a, and the flux density near the SE corner of the deer lick was 54×10^{-3}

$L/m^2 \cdot a$, indicating that the terrestrial flux can in this instant be almost 10-fold larger than the surface aquatic flux (Figure 3).

The total flux from creek stations 1–13b on July 21 was 137 L/a, and the total flux from the deer lick on July 20, using the chamber data, was 3.5 L/a. The average flux density from 24 measurements along the two transects connecting the creek to the deer lick (Figure 3) was $0.18 \pm 0.61 \times 10^{-3}$ $L/m^2 \cdot a$ on July 20. The wetland area to attribute this flux density to is arbitrary. Initially, we digitized the whole lowland from the creek edge on the west to the ridge on the east, from station 1 to 13b, to obtain an area of 44 000 m^2, omitting the 360 m^2 area of the bare deer lick. This terrestrial area is five times larger than the whole creek surface of 8500 ± 50 m^2. The flux density of 0.18×10^{-3} $L/m^2 \cdot a$ multiplied by this lowland area gives a flux of 7.9 L/a and combined with the 3.5 L/a of the lick gives a total terrestrial flux of 11.4 L/a. This overall aquatic–terrestrial partition is 92% aquatic and 8% terrestrial. Since the transect data are most relevant to the wetland area from station 6 to station 13, we digitized this area separately. The area from the creek to the ridge and bounded by station 6 on the north and station 13 on the south is 28 400 m^2, giving a terrestrial flux from this smaller area of 5.1 L/a and including the lick for a total flux of 8.6 L/a. The comparable area of the creek from station 6 to station 13 is 2540 m^2 and has a flux of 50 L/a. This suggests that the aquatic–terrestrial partition is 83% aquatic and 17% terrestrial.

The aquatic flux accounts for up to 92 ± 1% of the He released from this flood plain, and the terrestrial flux can account for up to 17%. This indicates that these adjacent aquatic and terrestrial flood plain areas that exhibit anomalous He in water, groundwater, and soil gas have deep groundwater preferentially exiting at the aquatic location. However, the data from this investigation to partition deep groundwater indicates that it is possible to have larger terrestrial than aquatic discharges. In 1991, a relatively dry year, we calculated an average flux of 270 L/a, assuming the peat was unsaturated and there was no effect of tortuosity. We considered this an overestimate of the flux. This is 1.6 times larger than the total flux of 170 L/a from the creek on July 21, 1993, and 77 times larger than the average flux from the deer lick measured in this study. Not surprisingly, the flux varies depending on the moisture status of the overburden and the hydrological budget of the watershed.

Conclusions

Use of the gas flux chambers on the terrestrial site was limited due to flooding. Since most flood plains or riparian wetland areas spend some portion of the year submerged, this makes direct flux measurements difficult if not impossible. The ping-pong ball method is excellent at determining the He concentrations in saturated soil. A calculated flux, however, is directly dependent on the transport coefficient. This coefficient varies over several orders of magnitude depending on the air-filled porosity, making it necessary to measure the soil dry bulk density, particle density, and moisture content.

He flux densities, measured directly on the deer lick in 20 chambers, varied from 0 to 54×10^{-3} $L/m^2 \cdot a$ giving a maximum flux of 8.3×10^{-4} L/a in the southeast corner of the lick. The average flux density from the entire deer lick was 9.75×10^{-3} $L/m^2 \cdot a$, producing an average He flux of 3.5 L/a over this 359.4 m^2 area. Helium flux calculations and measurements made on this lick over 3 years range from 3.5 to 270 L/a. The short stake transects indicate that He diffuses from isolated locations, primarily at the deer lick, rather than along major lineaments. This suggests that pipe flow occurs at fracture infill weaknesses rather than uniform flow along a planar fracture at this site. Fluxes measured at long ping-pong ball stakes varied from 0.27 to 277 $L/m^2 \cdot a$ depending on the season and the moisture conditions of the site.

Four creek locations showed elevated surface concentrations, and two of these were at points where the creek direction was diverted. If we join these two locations on the creek with the hot spot of the deer lick, the line follows a NW–SE direction, coincident with lineament directions. Although the creek He flux densities are generally higher than the wetland fluxes, one position in the SE corner of the deer lick produced similar fluxes early in the season to those measured at station 6 in the creek.

For the assessment modeling of the transport of radioactive and chemically hazardous elements from a vault located deep in the geosphere to the biosphere, we have used a ≤90% aquatic and a <10% terrestrial partition. Based on this study of one location, we suggest an annual partitioning of ≥80% aquatic and <20% terrestrial correctly models the potential for contaminants to exit from the geosphere to a terrestrial or semiterrestrial area.

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