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Regional survey for He anomalies in Canadian Shield lakes: sources of variation and implications for nuclear fuel waste management

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Abstract—Bottom-water He concentration (=[He]) was measured in the late summers of 1989, 1990 and 1991 in the waters of lake basins at the Experimental Lakes Area in northwestern Ontario, Canada. A total of 32 basins were sampled every year. Helium concentrations were relatively stable from year to year, exhibiting a mean coefficient of variation of 34%. Total [He] ranged from values below the atmospheric equilibrium concentration of 47 nl He/l H_2O to a maximum of 5364 nl/l measured in Lake 625 in 1991. Total [He] exhibited a two-phase distribution, with a large subpopulation of basins having only modest He enrichment (geometric mean G.M. = 77 nl/l, geometric standard deviation G.S.D. = 1.65, n = 39), and a small subpopulation of basins, including Lakes 625, 634 and 615, with large He anomalies (G.M. = 692 nl/l, G.S.D. = 3.08, n = 6). Using hydrological, morphometric and physical data for each lake, bottom-water [He] was predicted. A model including lake order and lake width accounted for 22% of the total variance in [He]. These results support the hypothesis that excess He in lake-bottom-water originates with deep groundwater discharge via fractures in the underlying granite.

INTRODUCTION

THE CANADIAN concept for the permanent disposal of nuclear fuel waste suggests that a repository constructed at depth (500–1000 m) in plutonic rock on the Canadian Precambrian Shield will safely isolate hazardous radionuclides from the biosphere. The most likely and serious breach of containment will arise if the radionuclides become dissolved in groundwater and are transported by advective processes to the surface (WUSCHKE *et al.*, 1981). An accurate description of discharge zones is a prerequisite to the site-specific modeling of environmental impacts that could result in the future. Consequently, it is important that methods for locating and characterizing deep-groundwater discharge points on the Shield are developed and tested.

Deep groundwater in Precambrian Shield regions tends to evolve toward characteristic brines (JOHNSTON, 1982; FRAPE *et al.*, 1984; PEARSON, 1987; GASCOYNE *et al.*, 1987). These brines are generally of great age. Associated with the age and salinity is a tendency for the brines to contain large amounts of dissolved He gas. This gas is derived from the *in situ* alpha-particle decay of natural U and Th isotopes (ANDREWS, 1987). Because He is far more mobile in fracture fluids than in unfractured rock, the diffusion and advection of fracture fluids can supply He, in excess of that expected from equilibrium with the atmosphere, to surface water bodies.

Because the primary sources of excess He (He_{ex}) in groundwater are the U- and Th-series radioactive decay chains, it has been proposed (CLARKE and KUGLER, 1973) that He_{ex} in groundwater can be used to identify economically valuable U deposits. TORGERSEN and CLARKE (1978) claimed to have detected such an ore body based upon the measurement of He_{ex} in lake bottom-water. Although studies in areas with known U mineralization (CLARKE *et al.*, 1977; DYCK and TAN, 1978) have correlated the locations of He_{ex} in lake water with the locations of U mineralization, other studies (DYCK and DA SILVA, 1981; GREGORY and DURRANCE, 1987; DYCK and CAR, 1987; GASCOYNE *et al.*, 1992; STEPHENSON *et al.*, 1992) indicate that He anomalies in lake water and soil gas are primarily associated with deep-seated fault and fracture zones.

In this paper the distribution of He anomalies in lakes located on a granite pluton at the Experimental Lakes Area (ELA) in northwestern Ontario is evaluated. Specifically, hypotheses concerning the association of elevated He concentrations (=[He]) in lake bottom water with lake basin order, morphometry and altitude were tested. The magnitude of interannual variations in the He anomalies was also measured, and the results of other lake surveys for He anomalies were reviewed in order to consider regional sources of variation.

STUDY AREA

The Experimental Lakes Area is located at 93°30′–94°00′W, 49°30′–49°45′N in northwestern Ontario (Fig. 1), ~50 km east of Kenora. The lakes are described in detail elsewhere (CLEUGH and HAUSER, 1971; ARMSTRONG and SCHINDLER, 1971; BRUNSKILL *et al.*, 1971). SANBORN-BARRIE (1987) described the geology of the region. Briefly, the area is underlain

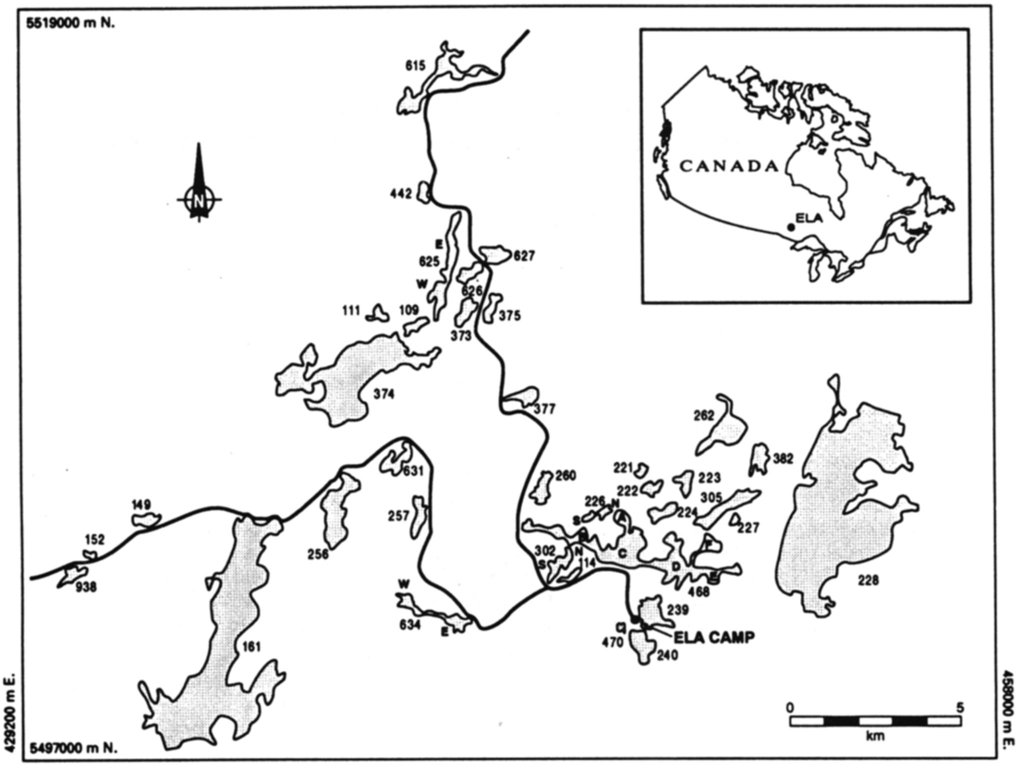

FIG. 1. The Experimental Lakes Area in northwestern Ontario, Canada, showing the locations and numbers of sampled lakes.

by the Dryberry batholithic complex, an Archean granitoid terrain comprising several batholiths, stocks and plutons occupying ~2500 km^2 (BLACKBURN, 1981). The immediate study area lies primarily on plutonic granites described by SANBORN-BARRIE (1987) as ranging from equigranular granite and granite pegmatite ± granodiorite to K-feldspar megacrystic granite ± granodiorite. The study area is situated on the south lobe of a massive pink granodiorite batholith (DAVIES and PRYSLAK, 1967). Complex exposures of older sedimentary, volcanic and intrusive rocks are found 10–30 km east, south and west of the study area (BRUNSKILL and SCHINDLER, 1971). The Experimental Lakes Area generally lies at an altitude of 360–380 m above sea level, and relief in the area does not usually exceed 80 m (BRUNSKILL and SCHINDLER, 1971). In most of the study area, the strongly sheared, jointed and faulted bedrock controls local drainage systems, and many lakes lie in fault basins (BRUNSKILL and SCHINDLER, 1971). There are >1000 lakes, ranging in maximum depth from <2 to 175 m and in area from <1 to >1300 ha. The hydrology of the lakes is dominated by streamflow, precipitation onto the lake surface, and direct runoff across the shoreline boundary (NEWBURY and BEATY, 1980). Groundwater seepage is generally of little importance because of the shallowness of the surficial deposits and the generally low relief of these bedrock-controlled basins (NEWBURY and BEATY, 1980; KENNEDY, 1974). Most of the lakes have water renewal times of <10 a (NEWBURY and BEATY, 1980).

METHODS

Study lakes were chosen to lie along two transects running north–south and west–east, respectively (Fig. 1), as determined by the locations of access roads. Lakes were sampled in mid-August 1989, 1990 and 1991, at their deepest points. Multi-basin lakes (Lakes 226, 302, 468, 625 and 634) were sampled at the deepest point in each major basin. Table 1 lists the lakes visited and the years in which they were sampled.

Water samples were collected using either a peristaltic pump equipped with plastic hose line and a sampling stirrup (STEPHENSON *et al.*, 1992) at the bottom end, or a Wildco van Dorn water-sampling bottle. The choice of equipment depended largely upon the water depth at the sampling station. The peristaltic pump was most efficient to depths of 20 m, whereas the van Dorn bottle was most efficient at greater depths. A comparison of samples collected with both equipment types at a depth of 175 m in Lake 228 showed no significant differences in measured [He]. All water samples were collected from as close to the sediment–water interface as possible without including sediment in the sample. Water samples were stored in glass bottles with a 50-ml air headspace. Bottles were sealed with steel crown caps. STEPHENSON *et al.* (1992) give details of sampling and analytical procedures so only a brief outline follows.

Helium gas is sparingly soluble in water, so it preferentially partitions into air when a gas phase is present. In equilibrium with the atmosphere (5240 nl He/l air), surface waters at 10°C contain 47 nl He/l water. Excess He trans-

Table 1. Measured He concentrations (nl/l) and $[Cl_b^-/Cl_s^-]$* in the study lakes

	Dissolved [He]						
Lake	1989	1990	1991	Mean†	S.D.‡	CV§	$[Cl_b^-/Cl_s^-]$
109	45	43	89	59	26	44	0.90
111	0	—	52	21	30	141	6.62
114	69	39	102	70	32	45	1.17
149	33	—	73	53	28	53	1.01
152	103	—	145	124	30	24	1.11
161	88	—	—	88	—	—	0.95
221	84	64	97	82	17	20	0.54
222	156	71	160	129	50	39	0.78
223	194	115	153	154	40	26	1.38
224	63	41	96	67	28	41	1.19
226N	93	57	96	82	22	27	1.41
226S	78	76	86	80	5	67	1.38
227	51	0	0	17	29	173	1.82
228	146	—	—	146	—	—	1.02
239	105	70	88	88	17	20	1.10
240	148	119	138	135	15	11	1.07
256	38	—	54	46	12	25	0.98
257	75	—	108	91	23	26	1.11
260	49	65	94	69	23	33	1.21
262	125	86	74	95	26	28	1.13
302N	65	70	80	72	8	11	—
302S	65	59	39	54	14	25	1.23
305	135	116	128	126	10	8	1.17
373	49	55	92	65	23	35	1.03
374	178	—	216	197	27	14	0.83
375	110	68	116	98	26	27	0.92
377	130	81	101	104	25	24	1.64
382	75	49	91	72	21	30	1.11
442	317	243	311	290	41	14	1.23
468A	127	39	157	108	61	57	1.02
468B	117	128	144	130	13	10	1.18
468C	68	48	71	62	13	20	1.02
468D	58	49	74	60	13	21	1.57
468E	57	33	77	56	22	40	0.87
468F	61	49	92	67	22	33	1.10
470	33	—	—	33	—	—	2.35
615	571	—	—	571	—	—	1.02
625E	1240	1142	1233	1205	55	5	2.43
625W	4427	4127	5364	4639	645	14	1.38
626	95	60	106	87	24	28	0.26
627	183	145	179	169	21	12	1.15
631	65	—	101	83	26	31	0.93
634E	—	166	—	166	—	—	—
634W	978	357	472	602	330	55	1.12
938	35	—	72	54	26	49	1.02

* Ratio of bottom-water to surface-water $[Cl^-]$. † Mean [He] for 1989–1991. ‡ Standard deviation. § Coefficient of variation (%). The mean [He] in the six outlier basins is underlined for clarity.

ported with groundwater into the hypolimnion of a lake and sampled into a sealed bottle will equilibrate with the headspace gas, resulting in an increased [He] in the headspace. By measuring the equilibrium [He] in the headspace gas, it is possible to calculate the original aqueous [He] in the water sample. BUTT and GOLE (1984) provided an equation and constants for this calculation:

$$He_w = (V_{hs}/V_w) \cdot (He_{hs} - He_a) + (\beta \cdot He_{hs}) \quad (1)$$

where He_w = actual [He] in water (nl He/l H_2O), He_{hs} = measured [He] in headspace (nl He/l air), He_a = concentration of He in air (5240 nl He/l air), V_{hs} = headspace gas volume (ml), V_w = volume of water sample (ml), and β = Bunsen solubility coefficient of He (0.00897).

Helium concentrations in headspace gas were measured using a Veeco MS-18AB mass spectrometer equipped with an adjustable inlet valve, and a cryogenic loop packed with activated charcoal, immersed in liquid nitrogen to remove water and other condensable vapors. Details of the analytical protocol are given by STEPHENSON *et al.* (1992).

In addition to water samples for He, samples for the determination of Cl^- were collected from the surface (grab sample) and bottom (peristaltic pump or van Dorn samples) waters of each lake in 1989. Chloride concentrations in these water samples were analyzed using a Dionex QIC anion chromatograph. In addition, bottom-water samples were collected from each lake in 1989 for the determination of dissolved ^{222}Rn concentrations by liquid scintillation counting, as described by STEPHENSON *et al.* (1992).

To explore the relations between bottom-water [He] and possible physical controlling factors, a combination of principal components analysis (PCA) with varimax rotation, and stepwise multiple regressions was used, using both principal component scores and the original variables to predict bottom-water [He] from physical parameters describing the lakes. Principal components analysis was used to reduce the dimensionality of the data describing each lake. The reduced variables (principal components) represent the original variables in the sense that they are linear combinations of the original variables, and are correlated with them at specified levels (Harris, 1975). Rotations of the principal components are used to modify the distribution of variance from the original data onto the principal components. Varimax rotations tend to polarize the loadings so that only a few variables correlate strongly with each principal component, simplifying their interpretation. Principal components have the additional advantage of being statistically orthogonal to each other, satisfying one of the major constraints of multiple regression analysis (Harris, 1975). Comparing the results of the multiple regression analysis using the original variables with those using the principal components simplifies their interpretation and helps to identify errors or bias arising from the lack of statistical independence in the original variables.

Parameters selected for PCA and multiple regression included lake order (total number of lakes upstream of the study lake outlet), lake altitude, lake depth, watershed area, lake length and lake width. Lake order, altitude and watershed area are all potential predictors of the location of the lake within the regional aquifer. Low-order high-altitude lakes with small watershed areas may provide recharge to the aquifer, whereas high-order lakes at low altitude with large watershed areas may receive groundwater discharge. Very deep lakes may be more likely to receive deep groundwater inputs. Long or wide lakes may lie in fracture-controlled basins, and consequently may be more likely to receive deep groundwater. Data for watershed area and lake order were obtained from an unpublished source (G. McCullough, Freshwater Institute Science Laboratories, Winnipeg, Manitoba, pers. commun. 1992), or from 1:50,000 scale topographic maps (Energy Mines and Resources Canada sheets 52 F/12, 52 F/13). Lake altitude, lake length and lake width were also measured from 1:50,000 scale maps. Lake depth was directly measured during the surveys using a Lowrance X-30 portable depth sounder. All variables were $\log_{10}$-transformed prior to the regression, to normalize distributions. Because of the poor predictive power of the available variables, a relatively weak probability criterion ($p < 0.2$) was used for inclusion of variables in the model. Statistical calculations were performed on a personal computer using commercially available software (SYSTAT 4.0).

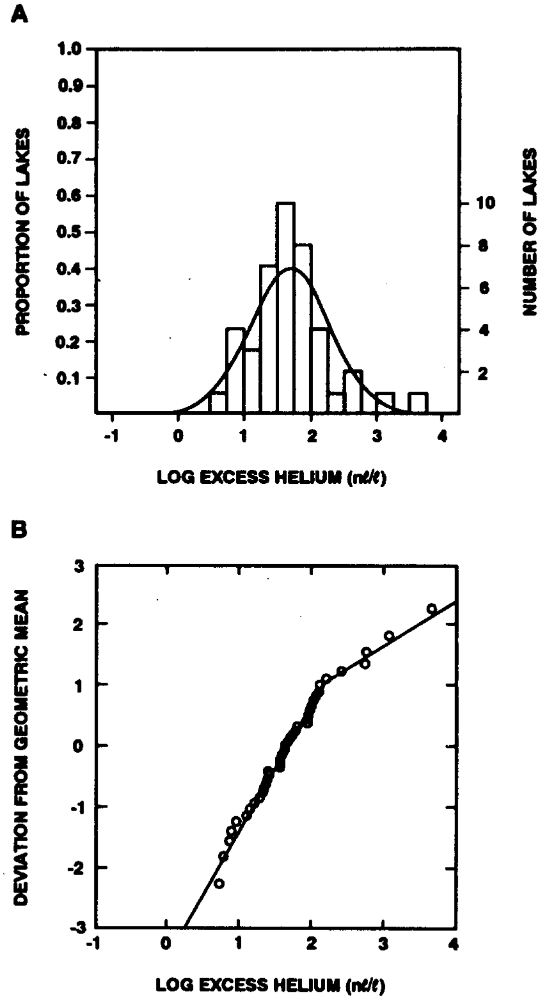

Fig. 2. (A) Histogram showing the distribution of excess He concentrations recorded in 46 lake basins. A normal distribution with G.M. = 102 nl/l and G.S.D. = 2.61 is superimposed. (B) Probability plot showing the distribution excess He concentrations recorded in the lakes. A straight line on this plot indicates a normal distribution. The y-axis indicates G.M. ± G.S.D. The data are best fitted by two lines, with a break between the subpopulations at ~180 nl/l.

RESULTS

The three annual lake surveys (Table 1) showed that although some lakes consistently have high [He] in bottom water, most approach the background (atmospheric equilibrium) concentration of 47 nl/l. Coefficients of variation within lakes are generally low, and do not change with increasing mean values. The annual data were subsequently pooled to obtain a 3-a mean [He] for each lake, and population statistics were calculated on the basis of these values.

Population statistics calculated for the 46 basins show that the distribution of [He] can be approximated by a truncated lognormal distribution (Fig. 2A) with a geometric mean (G.M.) of 105 nl/l and a geometric standard deviation (G.S.D.) of 2.58. However, an alternative presentation of the same data

Table 2. Principal components analysis of physical parameters describing the ELA lakes: rotated loadings of original variables on principal components

Variable	PC1	PC2	PC3	PC4	PC5
LO*	<u>0.957</u>	0.128	0.112	0.093	0.140
WA†	<u>0.916</u>	0.067	0.215	0.148	0.235
ALT‡	−0.124	<u>−0.969</u>	−0.100	−0.168	−0.086
DEP§	0.229	0.114	<u>0.919</u>	0.196	0.227
WID‖	0.158	0.188	0.188	<u>0.933</u>	0.183
LEN¶	0.287	0.102	0.240	0.201	<u>0.899</u>

*Lake order. †Watershed area. ‡Lake altitude. §Lake depth. ‖Lake width. ¶Lake length. All data $\log_{10}$-transformed prior to analysis. Loadings >0.5 are underlined for clarity.

Table 3. Stepwise multiple regression analysis of bottom-water [He] using PC1, PC5, PC4 and PC2, representing physical parameters of the lakes

Variable	Coefficient	S.E.	t	p
Intercept	2.016	0.058	34.89	<0.001
PC1	0.111	0.058	1.90	0.065
PC5	0.097	0.058	1.66	0.105
PC4	0.097	0.058	1.66	0.105
PC2	0.085	0.058	1.46	0.153

Analysis of Variance

Source	Sum of Squares	DF	Mean Square	F	
Regression	1.684	4	0.421	2.802	0.038
Residual	6.008	40	0.150		

$n = 45$ $r = 0.468$ $r^2 = 0.219$

All data $\log_{10}$-transformed prior to analysis.
The final regression equation has the form: $\log_{10}[He] = 2.016 + 0.111(PC1) + 0.097(PC5) + 0.097(PC4) + 0.085(PC2)$.

Table 4. Matrix of Pearson correlation coefficients for bottom-water [He] and physical parameters describing the study lakes

Variable	[He]	DEP	ALT	WAT	LO	WID
DEP	0.308					
ALT	−0.298	−0.283				
WAT	0.321	0.494	−0.247			
LO	0.395	0.390	−0.279	0.927		
WID	0.365	0.455	−0.393	0.378	0.310	
LEN	0.388	0.542	−0.269	0.559	0.462	0.462

Variable codes are as in Table 2. All data $\log_{10}$-transformed prior to analysis.

shows that two subpopulations may be present (Fig. 2B). The larger subpopulation, comprising 39 of the 45 basins sampled, has a G.M. [He] of 77.4 nl/l (G.S.D. = 1.65), whereas the remaining six basins have a G.M. [He] of 691.8 nl/l (G.S.D. = 3.08).

The PCA of six variables describing the physical attributes of the lakes results in the retention of five principal components (Table 2) that are readily interpreted through their correlations to the original variables. Briefly, the first principal component (PC1) represents lake order and watershed area, the second principal component (PC2) represents lake altitude, and PC3–PC5 represent lake depth, lake width and lake length, respectively.

A multiple regression analysis of [He], using the principal component scores as predictor variables, resulted in the inclusion of four PCs in a model (Table 3), accounting for 21.9% of the variation in bottom-water [He]. These were PC1 (lake order, watershed area), PC5 (lake length), PC4 (lake width) and PC2 (lake altitude). From the PC loadings (Table 2) and regressions (Table 3), it appears that bottom-water [He] increases with lake order, watershed area, lake length and lake width, and decreases with altitude.

The multiple-regression analysis using the original $\log_{10}$-transformed data as predictor variables included two predictor variables in a model predicting 22.2% of the variance in bottom-water [He]. Correlations between several of the predictor variables, particularly watershed area and lake order (Table 4), preclude their simultaneous use in regression models. Consequently, the optimal model included only lake order and lake width (Table 5). Figure 3A and 3B shows the correlations between lake order and bottom-water [He], and lake width and bottom-water [He], respectively, whereas Fig. 3C shows the correlation between the combined predictors, lake order and lake width, with bottom-water [He].

Measurements of Cl^- in bottom and surface water were made in all basins in 1989. The ratio of bottom-water to surface-water Cl^- concentrations ($[Cl_b^-/Cl_s^-]$) was generally near unity except in a few basins. Although these basins (Table 1) included those that demonstrated high [He], they also included several basins having little or no excess He. An attempt was made to measure $[^{222}Rn]$ in bottom water in 1989. No detectable (>7 Bq/l) ^{222}Rn was found in any basin during this survey.

Vertical He profiles were measured in several lakes during the three survey years (Fig. 4A–D). These profiles show that the He_{ex} is coming from the sediments. Measurements of [He] in the sediments of several basins having He_{ex} in bottom-water indicate that [He] increases by several orders of magnitude within 2 m of the sediment–water interface, demonstrating the existence of strong diffusive and advective fluxes of He_{ex} to the hypolimnetic water column in these lakes (STEPHENSON *et al.*, in prep.).

Table 5. Stepwise multiple regression analysis of bottom-water [He] using lake order and lake width

Variable	Coefficient	S.E.	t	p
Intercept	2.003	0.121	16.49	<0.001
LO	0.268	0.123	2.18	0.035
WID	0.336	0.179	1.88	0.068

Analysis of Variance

Source	Sum of Squares	DF	Mean Square	F	
Regression	1.704	2	0.852	5.976	0.005
Residual	5.988	42	0.143		
$n = 45$ $r = 0.471$	$r^2 = 0.222$				

Variable codes are as in Table 2. All data $\log_{10}$-transformed prior to analysis. The final regression equation has the form: $\log_{10}[He] = 2.003 + 0.268(LO) + 0.336(WID)$.

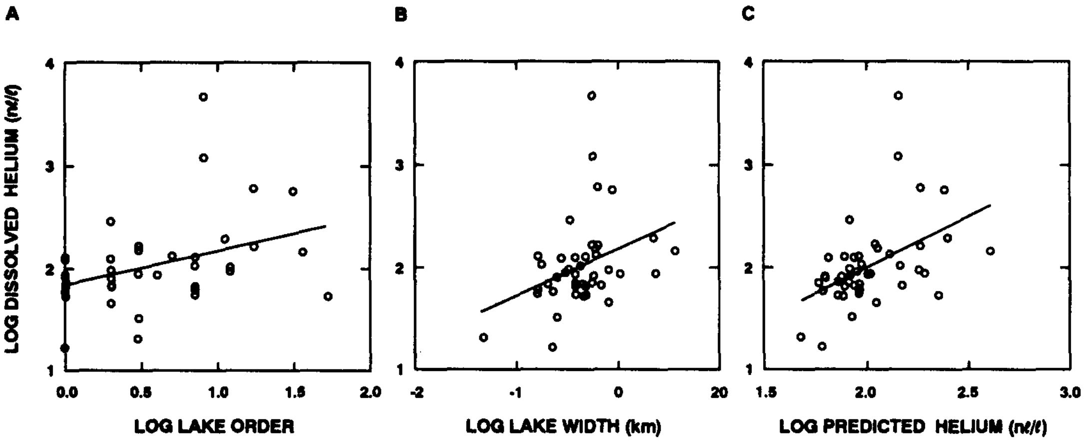

FIG. 3. Relations between bottom-water He concentration and: (A) lake order; (B) lake width; and (C) the combined predictors (lake order and width) from the multiple regression model.

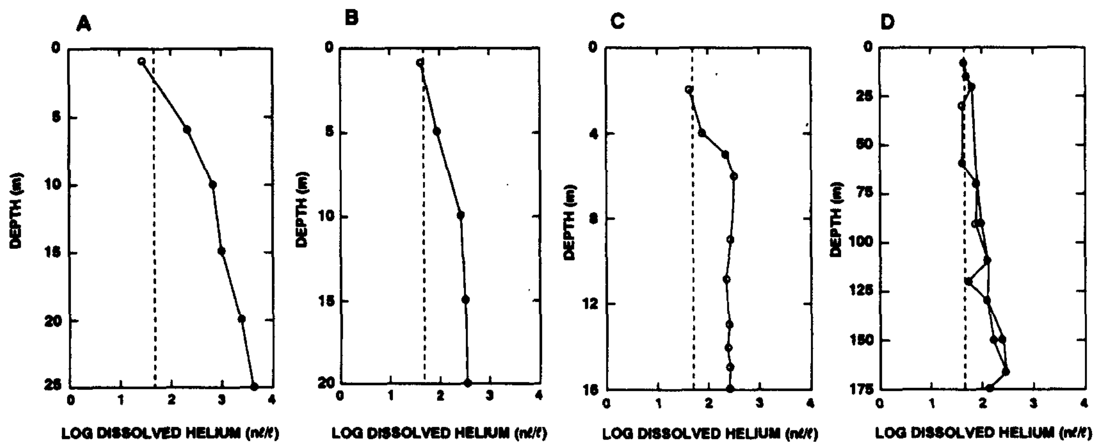

FIG. 4. Vertical He profiles measured in the lakes. (A) Lake 625 West; (B) Lake 634; (C) Lake 442; (D) Lake 228 (Teggau). Also presented in the last panel are data (●) from TORGERSEN and CLARKE (1978).

DISCUSSION

Within the study lakes, bottom-water [He] varied from 0 to 5364 nl/l. Because dissolved gases in most temperate zone lakes can be assumed to equilibrate with the atmosphere at spring and fall overturn, and He is inert, these lakes are expected to have aqueous [He] of ~47 nl/l. Little variation with depth is expected. The results presented here, and those of other workers (CLARKE *et al.*, 1977; TORGERSEN and CLARKE, 1978; CAMPBELL and TORGERSEN, 1980; DYCK and TAN, 1978; DYCK and DA SILVA, 1981;

DYCK and CAR, 1987) show that this is often not the case.

CLARKE *et al.* (1977), suggested that He_{ex} in lake water indicates the presence of significant U mineralization in the adjacent bedrock. TORGERSEN and CLARKE (1978), argued that a He anomaly found in Teggau Lake (Lake 228) identified such an ore body. They hypothesized the presence of a mass of 10^4 kg of U beneath Teggau Lake. They also recognized, however, that a fracture network could deliver He from a diffuse source to the lake. Based on $^3He/^4He$ data, CAMPBELL and TORGERSEN (1980), suggested that the slight enrichment of He found at a depth of 14–16 m in Lake 120 was due to air injection of atmospheric gases during the overland flow of cold meltwater in spring and density flow in the lake. While we accept this example of He enrichment by air injection in lake bottom waters, we contend that the simplest explanation for a large [He_{ex}] is the seepage of He-enriched deep groundwater from major fracture systems underlying water bodies, as suggested by DYCK *et al.* (1978), DYCK and CAR (1987) and STEPHENSON *et al.* (1992). GREGORY (1987), GREGORY and DURRANCE (1987) and GASCOYNE *et al.* (1992) have also argued strongly that the presence of He anomalies in surface waters or soil gases in plutonic terrain is an indicator of regional groundwater circulation toward major fracture zones.

The measurements of Cl^- in lake bottom and surface-water in this study generally show little variation from the ratio of 1.0 expected for a conservative tracer. Several basins, however, including some with high [He], have [Cl_b^-/Cl_s^-] > 1. While this may be coincidental, it suggests a deep groundwater origin for the He_{ex}, because high [He] is associated with groundwater brines found at depth on the Canadian Shield (BOTTOMLEY *et al.*, 1984). Groundwater samples from the Lac du Bonnet batholith in southeastern Manitoba, which is similar in its mineralogical composition to the Dryberry batholith, show a strong correlation between [He] and [Cl^-] (BOTTOMLEY *et al.*, 1984). Some of the basins having [Cl_b^-/Cl_s^-] > 1 can be explained on the basis of their meromictic status (e.g. Lake 111) or high productivity (e.g. Lake 227). In these basins, the decomposition of organic detritus results in the diagenetic release of some Cl^-. Other basins exhibiting high [Cl_b^-/Cl_s^-], including Lake 377 and Lake 470, are not easily explained. To summarize, while it appears that lakes having high [He] may also have slight enrichments of Cl^- derived from groundwater in bottom water, other limnological processes can also result in similar enrichments. Thus, while high [Cl_b^-/Cl_s^-] supports the hypothesis that He_{ex} is derived from deep groundwater, Cl^- measurements alone cannot identify areas of deep groundwater discharge. STEPHENSON *et al.* (1992) working in the Boggy Creek catchment in southeastern Manitoba, reached a similar conclusion. There, Cl^- measurements were complicated by interference from high dissolved organic C concentrations, and by apparent plumes of Cl^--enriched water from household septic drainage fields.

Two meromictic basins (Lakes 111, 615) and one (Lake 227) that approaches a meromictic condition owing to its small size, sheltered location, depth and eutrophic state, were included in the present survey. Lake 615 was highly enriched with He_{ex} in both the monimolimnion and hypolimnion. The other two basins had bottom water [He] close to the atmospheric saturation level one year, and no detectable He in the other years. It is suggested that the intermittent mixing of water into the monimolimnion (Lake 111) or hypolimnion (Lake 227) as demonstrated by CAMPBELL and TORGERSEN (1980) for Lake 120, results in the occasional infusion of He into the bottom water. The monimolimnion is quickly restabilized by the oxidation and re-precipitation of Fe^{2+} at the chemocline (CAMPBELL and TORGERSEN, 1980). In subsequent years, when the monimolimnion is not invaded by fresh water or the hypolimnion is not completely mixed at overturn, the degassing (bubble ebullition) of CO_2, CH_4 or H_2S from the bottom-water results in the simultaneous sparging and loss of He.

The absence of Rn from lake bottom water suggests that the He anomalies do not identify near-surface U mineralization. Radon is a daughter isotope of ^{226}Ra in the ^{238}U decay chain. Owing to its short half-life, ^{222}Rn does not persist long in groundwater. For this reason, ^{222}Rn can identify local concentrations of U and its daughter isotopes if groundwater travel times are short. DYCK and TAN (1978) measured [^{222}Rn] as high as 8 Bq/l (mean = 0.4 Bq/l) in association with small [He_{ex}] (maximum = 33 nl/l, mean = 4.4 nl/l) in lake-bottom-water under winter ice at Key Lake in northern Saskatchewan, an area with substantial U deposits. In another study, DYCK and CAR (1987) recorded a massive He anomaly (26,770 nl/l) in the bottom water of Dop Lake, Northwest Territories, but found only traces of Rn (G.M. = 0.03 Bq/l, maximum = 4.1 Bq/l). They concluded that the He anomaly was caused by upwelling groundwater supplied by a major fracture system underlying the lake, and discounted the possibility of substantial U mineralization adjacent to the lake because of the very low [^{222}Rn]. The [He_{ex}] values reported here are orders of magnitude greater than those reported by DYCK and TAN (1978) and approach those reported by DYCK and CAR (1987), yet ^{222}Rn was consistently below the detection limit (7 Bq/l). Although the analytical technique used has limited sensitivity, it should be adequate to detect ^{222}Rn in lake water if U mineralization is adjacent and groundwater transit time is short. LEMIRE and GASCOYNE (1987) reported [^{222}Rn] of from <1 to 485 Bq/l (G.M. = 26 Bq/l; G.S.D. = 7.2) for groundwaters and springs in the Whiteshell and Atikokan Research Areas, in Manitoba and northwestern Ontario, respectively. Moreover, in the absence of He_{ex}, ^{222}Rn was detected in a creek (13 Bq/l) and a

Table 6. Reported maximum [He] for lakes on the Canadian Shield

Region	Lake	[He_{max}]*	Reference
ELA	Lake 625	5364	This study
	Teggau	306	TORGERSEN and CLARKE (1978)
	Lake 120	86	CAMPBELL and TORGERSEN (1980)
Labrador	McLean	112	CLARKE *et al.* (1977)
Central Ontario	McKay	77	P. RASMUSSON (pers. commun.)
Manitoba	Boggy Lake	2877	STEPHENSON *et al.* (1992)
	Boggy Creek	12,789	STEPHENSON *et al.* (1992)
Saskatchewan	Key Lake	92	DYCK and TAN (1978)
	Key Lake	96	DYCK *et al.* (1978)
Northwest Territories	Dop Lake	26,770	DYCK and CAR (1987)

*Maximum bottom-water [He] reported in the study referenced.

spring (21 Bq/l) discharging into Lake 228. These observations are interpreted as evidence of short-term groundwater transport through Ra-bearing media, resulting in elevated [^{222}Rn] in the groundwater, without sufficient groundwater residence time to accumulate significant [He_{ex}]. This is consistent with the interpretations of JACOBY *et al.* (1979) who used natural He and ^{222}Rn isotopes to study groundwater supply to the Colorado River.

Attempts to predict [He] from the physical characteristics of the lakes had limited success, resulting in a model that accounts for 22.2% of the total variation in bottom-water [He], based upon lake order and lake width. The results of the multiple regression using PC scores had similar ($r^2 = 0.219$) predictive power, but suggested that lake length and lake altitude are also predictors. This discrepancy may be due to the lack of statistical independence in the original variables, and the masking of effects by correlated variables. Based on the simple regression results using the original variables, lake order was significantly, but weakly correlated ($r^2 = 0.156$) with [He]. This is intuitively reasonable because high-order lakes occur well downstream in regional drainage systems and are likely to be sites of regional groundwater discharge. Conversely, headwater lakes tend to occur high up in the regional drainage systems, and may tend to be sites of regional groundwater recharge. Lake altitude was negatively correlated with [He] ($r^2 = -0.089$). This is also consistent with the idea that high-altitude lakes will be sites of regional recharge, whereas low altitude lakes will be sites of discharge. Lake length and width were also correlated with [He] ($r^2 = 0.151$ and 0.134, respectively). These results appear to be reasonable, because lake basins on the Precambrian Shield commonly occupy eroded areas adjacent to faulted zones in the bedrock (BOSTOCK, 1976; BRUNSKILL and SCHINDLER, 1971). Many lakes at the study area lie in fault basins (BRUNSKILL and SCHINDLER, 1971) and tend to be elongated. Despite their correlation, high values for both lake length and width may indicate the intersection of fracture lineaments within a lake basin. YIN and BROOK (1992) showed that the distance from a well site to the nearest fracture intersection (as determined from an analysis of aerial photographs) was a powerful predictor ($r^2 = 0.59$) of well yield in the crystalline rocks of the Georgia Piedmont and Blue Ridge area. GREGORY (1987) and GREGORY and DURRANCE (1987) showed that fractures exercise the dominant control on deep groundwater circulation, with associated He anomalies, in the Carnmenellis granite pluton of southwest England. Consequently, the associations between lake width, length and high [He] may be a result of the underlying structural geology.

Many unmeasured factors could exercise stronger influence upon the bottom-water [He]. Clearly, local variations in the U content of the granite will influence the flux of radiogenic He to the groundwater. Similarly, local variations in the intensity of fracturing and variations in fracture width or permeability may modify local fluxes of deep groundwater and He to lakes. It is beyond the scope of the present investigation, however, to attempt to measure these parameters on a regional scale.

Considering the [He] of lakes included within this survey as characteristic of a population of lakes distributed over a granite pluton, the distribution type and parameters of the population have been described (Fig. 2A, B). The lognormal distribution also provides a good descriptive model (G.M. = 56 nl/l, G.S.D. = 1.25) for the data of CLARKE *et al.* (1977) (Fig. 5). Other studies have also found He anomalies to be generally small and rare. A population of 25 central Ontario lakes had a bottom-water G.M. [He] of 41 nl/l (G.S.D. = 1.61) (P. Rasmusson, McMaster University, pers. commun.). Many of these lakes were undersaturated with He, possibly reflecting the sparging of hypolimnetic waters by gases of decomposition resulting from cultural eutrophication. DYCK *et al.* (1978) reported only 4 lakes of 99 surveyed near Key Lake in Saskatchewan to have [He] significantly greater than the atmospheric equilibrium concentration. This population had G.M. = 44 nl/l, G.S.D. = 1.10. The total [He] measured in

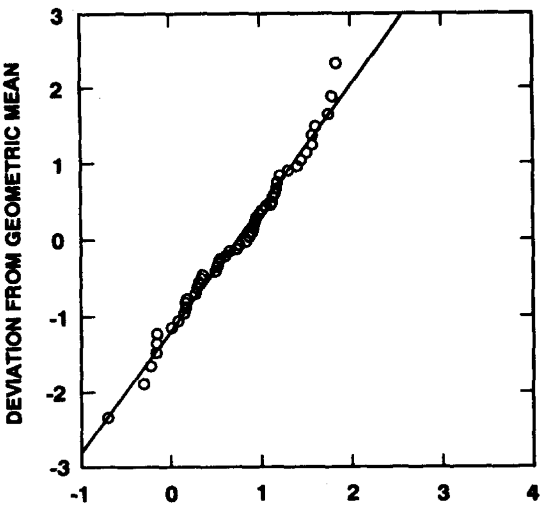

FIG. 5. Probability plot showing the distribution of [He] in Labrador lakes (CLARKE *et al.*, 1977). This population has G.M. = 56 nl/l, G.S.D. = 1.25.

lakes of the study area collectively form the most He-enriched population yet reported (G.M. = 105 nl/l, G.S.D. = 2.58), and include some of the highest values reported for individual lakes on the Canadian Shield (Table 6). Only Dop Lake in the Northwest Territories (DYCK and CAR, 1987) has a higher reported [He] than Lake 625. A site in Boggy Creek, Manitoba, also had a very high [He] (STEPHENSON *et al.*, 1992), but this site was very localized and lay directly over a fracture intersection with the creek.

Inspection of the data presented here (Fig. 2B) suggests that, rather than being log-linear, the ELA data contain a break point, with most of the values lying on one lognormal line, and a few very high values lying on another, independent line. Viewed this way, there may be two populations of lakes present at the study area: a large population of lakes with modest [He]; and a small population of lakes with very high [He], possibly situated above regional fractures with particularly high permeability or strong upward hydraulic gradients, resulting in the discharge of He-enriched groundwater through the lake sediments. This hypothesis is supported by vertical He concentration profiles measured in the sediments of a number of the lakes between 1989 and 1991 (STEPHENSON *et al.*, in prep.). In all of these cases, He concentrations increase with increasing depth, showing that He is entering these lakes through the sediments. Thus He surveys of this kind can be used to identify lakes receiving deep groundwater inputs, and an analysis of the population distribution of [He] in lake bottom-water may identify lakes situated over high permeability fractures. This technique is clearly relevant to the nuclear fuel waste management program, and should complement regional hydrogeological modeling during future site screening and evaluation programs.

SUMMARY

Measurements of dissolved [He] in lakes occurring on a granite pluton at the Experimental Lakes Area in northwestern Ontario show that most lakes contain little He_{ex} relative to the atmospheric equilibrium concentration of 47 nl/l. A few basins, outliers to the approximately lognormal distribution of [He] demonstrated by most of the population, exhibit very large He anomalies. We suspect that these basins are receiving deep groundwater inputs, enriched in both He and dissolved salts, from underlying fracture systems in the granite. We base this belief upon several lines of evidence:

(1) Although most lakes contain little He_{ex}, a few lakes are highly enriched with He. The most likely source of this He is the seepage of deep groundwater enriched with He from the radioactive decay of natural U- and Th-series isotopes.

(2) The absence of ^{222}Rn anomalies in conjunction with excess He shows that we are not detecting near-surface U mineralization. Thus, the seepage of He-enriched groundwater along a long flow path is implicated.

(3) Slight enrichments of bottom-water [Cl^-] relative to surface water concentrations occurring in lakes with He anomalies suggest a deep groundwater source.

(4) Relations between lake order and [He] show that high-order lakes are more likely to contain He anomalies than headwater lakes. Headwater lakes are likely sites of regional groundwater recharge, whereas high-order lakes occur lower in the regional topographic setting and occupy likely discharge areas. Similarly, lake length and width, which may identify geological lineament axes intersecting in the lake basin, are also predictors of bottom-water [He].

Based upon this evidence, it appears that regional He surveys can provide useful data to identify areas of regional deep groundwater discharge on the Precambrian Shield. This type of information may be useful for the future siting of a high-level nuclear fuel waste repository.

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