

Sedimentation Rates and Sediment Core Profiles of ^{238}U and ^{232}Th Decay Chain Radionuclides in a Lake Affected by Uranium Mining and Milling

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Radionuclide concentration profiles in sediment cores from three deep basins of Quirke Lake, Ontario, showed enrichment in the surficial layers related to the period of uranium mining in the watershed, based on sedimentation rates determined from a milling-related pH decline indicated by fossil diatoms. Recent sedimentation rates are 96 ± 7 to $185 \pm 14 \text{ g} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$. ^{210}Pb profiles could not be used to determine sedimentation rates due to loadings of ^{226}Ra and ^{210}Pb from mining and milling activities. Profiles of ^{238}U and ^{232}Th decay chain radionuclides showed an enrichment of 1–3 orders of magnitude in surficial sediments relative to background in deeper sediments. Radionuclide levels in surficial sediments exceeded those reported in lake sediments in uncontaminated systems and, for some radionuclides, approached or equalled levels in sediments contaminated with uranium and radium refining residues. Radionuclide activity ratios in the surficial layers of the Quirke Lake cores and in downstream sediments demonstrated the relative mobilities of the elements in the watershed. Net sediment loading rates for radionuclides in the three basins were $130\text{--}230 \text{ mg} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$ for U, $340\text{--}4000 \text{ Bq} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$ for other members of the ^{238}U decay chain, and $210\text{--}530 \text{ Bq} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$ for ^{232}Th and ^{228}Th .

Des profils de concentrations de radionucléides obtenus de carottes de sédiments prélevées dans trois bassins profonds du lac Quirke (Ontario) montrent un enrichissement des couches superficielles lié à l'exploitation de l'uranium dans le bassin hydrographique. Ce fait est basé sur les taux de sédimentation déterminés à partir d'une baisse du pH liée au broyage et révélé par les diatomées fossiles. Les taux de sédimentation récents varient de 96 ± 7 à $185 \pm 14 \text{ g} \cdot \text{m}^{-2} \cdot \text{a}^{-1}$. Les profils du Pb^{210} n'ont pu être utilisés pour déterminer les taux de sédimentation à cause des charges de Ra^{226} et de Pb^{210} provenant de l'exploitation minière et du broyage. Les profils des radionucléides U^{238} et Th^{232} dans la chaîne de désintégration montrent un enrichissement allant de 1 à 3 ordres de grandeur dans les sédiments superficiels par rapport à la concentration ambiante dans les sédiments plus profonds. Les teneurs en radionucléides des sédiments superficiels étaient supérieures à celles des sédiments lacustres de systèmes non contaminés et, pour certains radionucléides, légèrement inférieures ou égales aux niveaux signalés dans les sédiments contaminés par des résidus du raffinage de l'uranium et du radium. Les taux d'activité des radionucléides dans les couches superficielles des carottes du lac Quirke et les sédiments d'aval montrent les mobilités relatives des éléments dans le bassin hydrographique. Les taux de charge nets en sédiments de radionucléides dans les trois bassins variaient de 130 à $230 \text{ mg} \cdot \text{m}^{-2} \cdot \text{a}^{-1}$ dans le cas de l'U, de 340 à $4000 \text{ Bq} \cdot \text{m}^{-2} \cdot \text{a}^{-1}$ pour les autres membres de la chaîne de désintégration du U^{238} et de 210 à $530 \text{ Bq} \cdot \text{m}^{-2} \cdot \text{a}^{-1}$ dans le cas du Th^{232} et du Th^{228} .

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The uranium mining and milling industry is active in two regions of Canada — near Lake Athabaska, Saskatchewan, and near Elliot Lake, Ontario. These operations release natural radionuclides of the ^{238}U and ^{232}Th decay chains to aquatic and atmospheric environments, primarily from tailings disposal areas. Large amounts of acid, stable trace metals, and milling reagents are also released to the

environment by these operations. Once released, radionuclides are transported through environmental pathways and may impact radiologically on human receptors and natural biota.

In freshwater systems, transport of radionuclides is influenced to some degree by sorption onto particulate phases, precipitation reactions, and uptake by biota. Settling of these particulate forms results in a loading to the sediments. As well,

soluble forms of radionuclides may diffuse between lake water and bottom sediments. Sedimentation of radionuclides acts to reduce downstream export and thus moderates the impact of releases in downstream areas. Following the cessation of mining and milling and the reduction or termination of aqueous releases, these contaminated sediments may act as a source of radionuclides to the water column, resulting in long-term impacts.

^{210}Pb and ^{226}Ra of the ^{238}U decay chain have been widely measured in sediment cores to determine sediment accumulation rates in studies on nutrient cycling, trace element loading, particle reworking, and sediment diagenesis (Robbins and Callender 1975; de March 1978; Kipphut 1978; Robbins 1978; Evans and Rigler 1980a, 1980b; Carignan and Flett 1981; Cornwell 1985). Terrestrial and atmospheric loadings and particulate-solute partitioning of ^{238}U decay chain radionuclides were studied in Lake 239 (Experimental Lakes Area, northwestern Ontario; Brunskill and Wilkinson 1987) and in four lakes in the eastern Keewatin District, Northwest Territories (G. J. Brunskill, Freshwater Institute, Winnipeg, Man., pers. comm.), in investigations of radionuclide budgets in watersheds unaffected by the uranium industry.

This study was undertaken to determine accumulation rates and the degree of contamination by ^{238}U and ^{232}Th decay chain radionuclides in the sediments of Quirke Lake, a large Shield lake draining much of the uranium mining and milling district near Elliot Lake. Sedimentation rates were measured to relate the time scale of sediment contamination with the history of mining and milling in the basin. Sedimentation rates are also of concern in light of recent interest in examining the feasibility of deep lake disposal of uranium mill tailings. The burial of emplaced tailings with "clean" sediment after the cessation of mining and milling would result in a barrier to diffusion that would increasingly inhibit remobilization of radionuclides from the sediments back to the water column over the long term.

Lush et al. (1982) modelled the downstream transport, migration through food chains, and effective dose commitments resulting from releases of radionuclides from a hypothetical tailings mass on the shore of Quirke Lake. This model was developed in part as a decision-making tool for regulatory agencies in the management of radioactive releases to surface waters from uranium mines and mills. Model parameters were assigned values based on a review of the literature, rather than on any specific measurements in Quirke Lake or downstream environments. In their sensitivity analysis, the authors found that model predictions (radiation doses) were sensitive to sedimentation parameters in downstream watersheds. The present study provides data on sedimentation rates and net radionuclide accumulation rates for Quirke Lake that may be used in the calibration of such models. Current research is focussing on sediment-porewater distributions of radionuclides, radionuclide concentrations in water-column particulates, and on measurement of aqueous inputs and outputs of radionuclides in Quirke Lake. Results of this work will be published in the near future.

Sediment chronologies can be determined from ^{210}Pb profiles only in cases where the supply of ^{210}Pb to the sediment occurs at a constant rate, or if the ^{210}Pb supply is proportional to a varying sedimentation rate (Appleby and Oldfield 1983). These assumptions are almost certainly violated in Quirke Lake because uranium milling increases radionuclide loadings to the lake and these loadings vary in quantity and relative propor-

tions of radionuclides depending upon the intensity of mining and milling activity and the wastewater treatment technologies employed.

To overcome this problem, modern sedimentation rates in Quirke Lake were determined by analysis of fossil diatom and pollen profiles in sediment cores. Diatom communities respond to acid loadings by shifting to a more acidophilous species assemblage (e.g. Battarbee 1984; Charles 1985). The sulphuric acid leaching process and the production of pyritic tailings in uranium milling led to the acidification of Quirke Lake beginning around 1957. More recent neutralization of aqueous discharge has permitted a recovery from pH 4.5–5.5 in the early 1960s to pH 5.5–6.4 observed during this study. Diatoms in the core samples were analyzed and lake pH-core depth relationships determined, using approaches similar to those described by Delorme et al. (1984) and Dickman et al. (1984). In Quirke Lake, the decline in diatom-inferred pH was used as a marker for the onset of acidification in 1957, and the mass of more recent sediment accumulation to estimate modern sedimentation rates. Historic sedimentation rates representative of the century preceding the mid-1950s were estimated by analysis of the *Ambrosia* pollen rise in sediment cores (e.g. Huhn 1974; Kemp and Harper 1976; Kemp et al. 1978; Warwick 1980; McAndrews and Boyko-Diakonow 1987).

Study Area

Quirke Lake is a relatively large, deep waterbody (Fig. 1) in the headwaters of the Serpent River watershed draining into the North Channel of Lake Huron. The lake is situated on the Precambrian Shield, approximately 120 km west of Sudbury, Ontario, at 46°30'N, 42°35'W. It is the largest lake in the Serpent River system, with a surface area of 2040 ha, mean and maximum depths of 39 and 102 m, respectively, and a hydraulic detention time of ~5 yr (Snodgrass et al. 1983).

At water depths >3 m, sediments in the lake consist of a dark, fine-grained gyttja with a surficial orange-brown flocculent deposit over about 50% of the lake bottom. Sediments in the deep basins (≥ 98 m) are brown-black muds, overlain by ~10 cm of an orange-brown layer in the two western basins and by ~10 cm of a light brown deposit in the eastern basin (Fig. 1). We found the orange-brown layer to be rich in iron (unpubl. data). Releases of iron from the mining and milling of pyritic ores are the probable source, although post-depositional diffusion of iron upward in reducing porewaters and precipitation at the oxidizing interface in the surficial sediments (e.g. Carignan and Flett 1981; Cornwell 1985) may contribute to this phenomenon.

The first uranium mine in the watershed began production in 1955. By 1957, there were 11 operating mines in the system. Uranium production decreased in the early 1960s and increased in the late 1960s in response to market demand. There are presently several active and inactive mining and milling operations and tailings disposal sites located within the Quirke watershed. Mill process waters are acidic due to the sulphuric acid leaching process used to dissolve uranium from the ore during milling. This acidity favours the release of radionuclides from the ores. Uranium is removed from the acidic solution, and other dissolved constituents are precipitated by neutralization and discharged with the tailings to the tailings management area. Acidic conditions develop in the tailings due to pyrite oxidation, resulting in the dissolution of radionuclides and subsequent release to surface waters. Acidic conditions

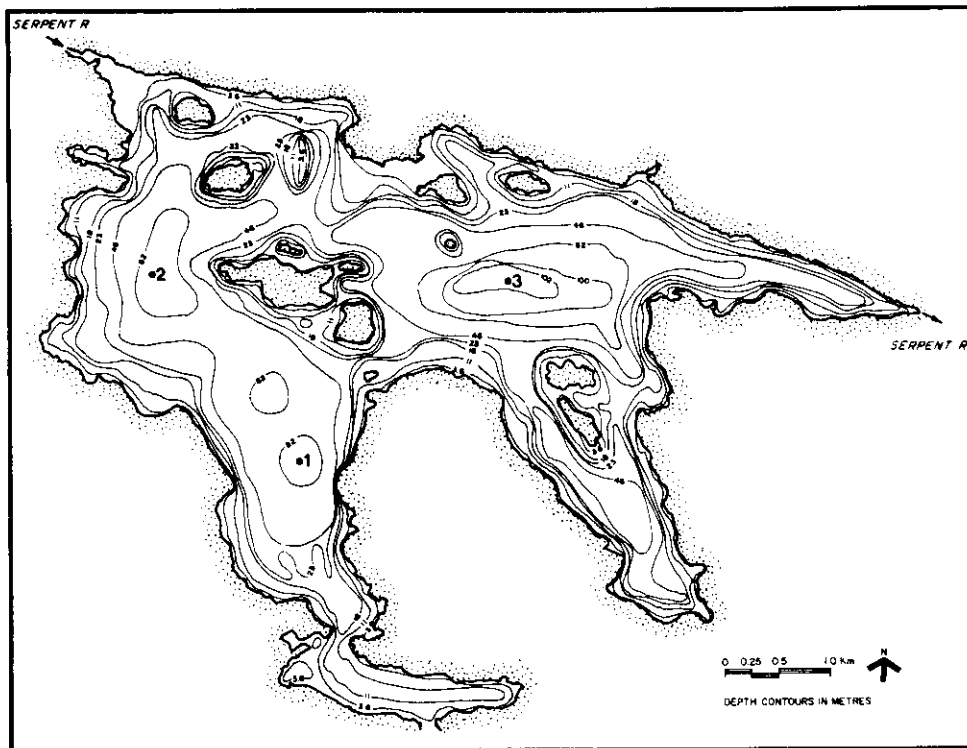


FIG. 1. Bathymetric map of Quirke Lake showing three sediment core sampling stations.

(pH 4.5–5.7) consequently occurred in Quirke Lake in the 1960s and 1970s, with a gradual increase in pH to 5.5–6.4 observed in 1984 (Ontario Ministry of the Environment, unpubl. data) apparently due to improvements in effluent treatment, neutralization of tailings seepage, and improved containment of leachates in new tailings structures over the last 10–15 yr.

Methods

Sediment cores were collected from Stations 1, 2, and 3 at depths of 98–101 m (Fig. 1) in October 1984 using a weighted KB corer outfitted with 4.7-cm (inside diameter) polycarbonate core tubes. Undisturbed cores about 30 cm long were collected with the aid of a CRT-type depth sounder to monitor core entry. Only cores exhibiting a flocculent layer at the sediment–water interface and a layer of overlying water were retained. Five cores were collected from each station — two cores for radionuclide analysis and one core each for diatom analysis, pollen analysis, and dry bulk density determinations.

Cores were sectioned fresh within 24 h of collection. Cores for pollen analysis were sectioned into 1-cm intervals from top to bottom. Before sectioning, horizontal laminations observed in the pollen cores were counted and measured. Cores for diatom analysis were sectioned into 0.2-cm intervals to a sediment depth of 2 cm, 0.5-cm intervals from 2 to 5 cm, and 1- to 3-cm intervals to a depth of 16 cm. Dry bulk densities were determined on 1.5-cm core sections after oven-drying. Duplicate cores for radionuclide analysis were sectioned into six 0.5-cm sections from the surface down to 3 cm; the duplicate sections from each depth were then combined to provide sufficient mass for analysis. One-centimetre-thick sections were taken from one of each duplicate set of radionuclide cores beginning at 3, 4, 6, 9, 14, and 19 cm. Samples for radio-

chemical analysis were frozen in plastic bags until analysis.

Samples for pollen analysis were first spiked with a known number of *Lycopodium clavatum* spores to permit accurate pollen density estimates (Stockmarr 1970). Fossil pollen was then concentrated from sediment samples with HCl, KOH, and acetolysis solution. Pollen grains and spores were counted until approximately 200 tree and shrub pollen grains per sample were obtained. Pollen was identified following McAndrews et al. (1973). Chronologically significant pollen horizons were expected to occur in more recent sediments; thus, every core section from the surface to 11–15 cm of depth was examined. Only three intervals between 11–15 cm and the bottom of each core were analyzed. Once analyzed, percentage abundance of pollen was used to define pollen zones in the cores based on the rise in weed pollen (*Ambrosia*, Gramineae, Chenopodiaceae, *Rumex*, *Plantago*) that can be related to the beginning of farming by European settlers in southern Ontario at about 1830–40 (McAndrews and Boyko-Diakonow 1987).

Core sections for diatom analysis were freeze-dried and blended and 0.05 g was then digested with a mixture of HNO₃ and H₂SO₄. Following washings with distilled water to remove acidity, diatom samples were quantitatively settled onto cover glasses in Battarbee chambers (Battarbee 1973) and then mounted on glass slides using Naphrax, a synthetic resin of high refractive index. At least 500 diatoms per sample were counted.

Several techniques have been developed to assess trends in acidification based on sedimentary diatoms using equations for inferring lake water pH. Problems and uncertainties associated with this methodology are discussed in Davis and Anderson (1984) and Charles (1985). In this study, we assigned each diatom species into one of five categories defined by Hustedt (1939): acidobiontic, acidophilic, indifferent, alkaliphilic, and alkalibiontic. Assignment of diatoms to these categories was

based on the work of Merilainen (1967) and Taylor (1986). Changes in the percent categories in a sediment core were used

$$B = \frac{\% \text{ indifferent} + 5 (\% \text{ acidophilic}) + 40 (\% \text{ acidobiontic})}{\% \text{ indifferent} + 3.5 (\% \text{ alkaliphilic}) + 108 (\% \text{ alkalibiontic})}$$

The predicted pH – sediment depth profiles were examined, and the historic pH decline was assumed to correspond to the acidification of Quirke Lake caused by the beginning of uranium milling in 1957 in the watershed.

Prior to analysis of radionuclide concentrations or activities, sediments were dried at 50°C and ground to ensure sample homogeneity. Samples for ^{226}Ra and Th isotope analyses were dissolved by KF-pyrosulphate fusion. Samples for total U, ^{210}Pb , and ^{210}Po analyses were digested in a hot, concentrated mixture of HNO_3 , HClO_4 , and HF, followed by a pyrosulphate fusion.

The methods used for radionuclide analysis are detailed in Chiu and Dean (1984) for U, Th isotopes, ^{226}Ra , and ^{210}Pb and in Smithson (1979) for ^{210}Pb . Total U in the solution was determined fluorometrically as its fluoride complex following fusion with NaF–LiF. Th isotopes were determined using high-resolution alpha spectrometry (HRAS). Prior to analysis, ^{234}Th was added to the digested sample as a tracer. Th was coprecipitated with PbSO_4 , isolated by solvent extraction, and then coprecipitated with cerous hydroxide. The precipitate was beta-counted for ^{234}Th to measure the overall yield and then ^{230}Th , ^{232}Th , and ^{228}Th measured by HRAS. ^{226}Ra was determined by HRAS using ^{133}Ba as a tracer. Ra was coprecipitated with BaSO_4 and the chemical yield measured by gamma counting of ^{133}Ba . ^{210}Pb was measured indirectly using ^{210}Bi . The sample solution was held for 30 d prior to analysis to permit greater than 98% ingrowth of ^{210}Bi . A carrier solution of stable Pb and Bi was added before digestion. Bi was then solvent extracted, precipitated as BiOCl , and ^{210}Bi measured by beta-counting. ^{210}Po was measured by HRAS using ^{208}Po as a tracer. Po isotopes were deposited on a nickel disc and the yield measured by recovery of the tracer. Radiochemical determinations were also carried out on one or two UTS reference standards, developed for the National Uranium Tailings Program, Department of Energy, Mines and Resources Canada, for every 15–20 sediment samples analyzed.

Radionuclide counting errors (1σ) were generally $\leq 12\%$ of total activity for all activity concentrations of ^{210}Pb and ^{210}Po and for ^{226}Ra activities $> 1 \text{ Bq} \cdot \text{g}^{-1}$. Errors were usually $\leq 25\%$ for Th isotopes at activities of $> 1 \text{ Bq} \cdot \text{g}^{-1}$. At lower activities, counting errors for ^{226}Ra and for Th isotopes generally ranged from 10 to 50%.

Results and Discussion

Sedimentation Rates

Two major pollen zones were found in each core (Fig. 2); Zone 3 is characterized as having $< 2\%$ weed pollen, while Zone 4 has $> 4\%$ (pollen zonation as described by McAndrews 1981). Since farming is not widespread in the North Channel region, it is assumed that southern Ontario is the major source of weed pollen in Quirke Lake sediments. Although ragweed (*Ambrosia*) is rare in the Quirke Lake area and northward, it is represented in the pollen record in lake sediments of the region

to reconstruct trends in lake water pH, following the Renberg and Hellberg (1982) index B approach:

(Huhn 1974). As in southern Ontario, the Zone 3/Zone 4 pollen zone boundary is assumed to be dated at about 1830–40 (McAndrews and Boyko-Diakonow 1987); thus, the overlying sediment represents ~ 150 yr of accumulation. The 3/4 boundary occurred at sediment depths of 12, 11, and 8 cm in Cores 1, 2, and 3, respectively. Sediments between the 3/4 pollen zone boundary and the diatom-inferred pH decline have accumulated between the beginning of southern Ontario farming and the beginning of uranium mining and milling in the watershed.

The beginning of lake water pH declines, based on diatom-inferred pH, occurred at a sediment depth of 6.5 cm at Station 1 and 4.25 cm at both Stations 2 and 3 (Table 1; Fig. 3). These depths are assumed to correspond to the 1955–59 period. Although the chronology of lake acidification is not precisely known, unpublished records indicate a relatively immediate pH decline following the start-up of uranium mining and milling and acidic conditions (pH 4.5–5.5) in Quirke Lake in the early 1960s. A date of 1957 for the pH decline probably has an uncertainty of ± 2 yr, since mining and stockpiling of pyritic ores began in 1955 and may have caused some acid loading at that time. Large-scale milling (and acid leaching) began in 1957. Based on a lake detention time of 5 yr, an additional 2-yr lag prior to lake acidification is also plausible.

The diatom-inferred pH values derived from the surficial sediment (Fig. 3) are within the present lake water pH range of 5.5–6.4. Furthermore, the inferred pH minima of ~ 5.4 –5.5 for Stations 1 and 2 correspond to reported minima of pH 4.5–5.5 for the early 1960s. However, the inferred pH minimum of 6.0 for Station 3 is at variance with this observation. Inspection of the diatom assemblages from Station 3 reveals a predominance of the pH-indifferent planktonic diatom *Cyclotella stelligera* in deeper sediments, which would tend to overestimate pH. The spatial variability of diatoms in sediments is mainly a function of basin morphometry, and according to the criteria of Earle et al. (1987), planktonic diatoms would tend to be overestimated and littoral diatoms underestimated at Station 3. Station 3 is located in the eastern portion of Quirke Lake and is somewhat isolated from the western portion containing Stations 1 and 2 by a chain of islands. This segregation of the two basins may account for the development of different diatom assemblages in each.

Sedimentation rates calculated from the fossil diatom and pollen data range from 96 ± 7 to $185 \pm 14 \text{ g} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$ (1.6 ± 0.2 to $2.4 \pm 0.2 \text{ mm} \cdot \text{yr}^{-1}$) for postmining sediments and from 34 to $64 \text{ g} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$ (0.31 to $0.55 \text{ mm} \cdot \text{yr}^{-1}$) for premining sediments (Table 1). Higher recent rates of sedimentation may be related to uranium mining and milling activities. Suspended solids levels may be high in runoff from disturbed areas, and precipitates formed from ferrous minerals dissolved by the milling process may accumulate in Quirke Lake sediments, as suggested by the iron-rich orange–brown deposits observed on the sediment surface. The whole-lake average sedimentation rate in Quirke Lake is probably lower than the accumulation rates found in the three deep basins, due to sed-

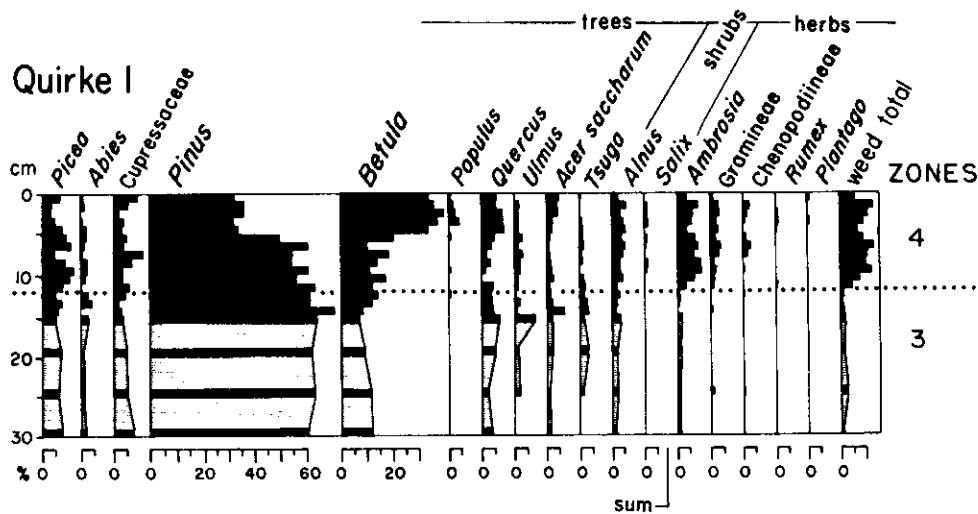


FIG. 2. Pollen diagram from core at Quirke Lake Station 1 showing percentages of main pollen types. Abundance of pollen expressed as percentage of total tree and shrub pollen. Dotted line represents the 3/4 pollen zone boundary (1835).

TABLE 1. Sedimentation rates in Quirke Lake on the basis of fossil pollen and diatoms, Stations 1, 2, and 3. Errors associated with sedimentation rates for postmining zone sediments are based on errors of ± 2 yr associated with dating of the acidification of Quirke Lake.

Zone	1		2		3	
	mm·yr ⁻¹	g·m ⁻² ·yr ⁻¹	mm·yr ⁻¹	g·m ⁻² ·yr ⁻¹	mm·yr ⁻¹	g·m ⁻² ·yr ⁻¹
Postmining	2.4±0.2	185±14	1.6±0.2	96±7	1.6±0.2	96±7
Premining	0.45	55	0.55	64	0.31	34

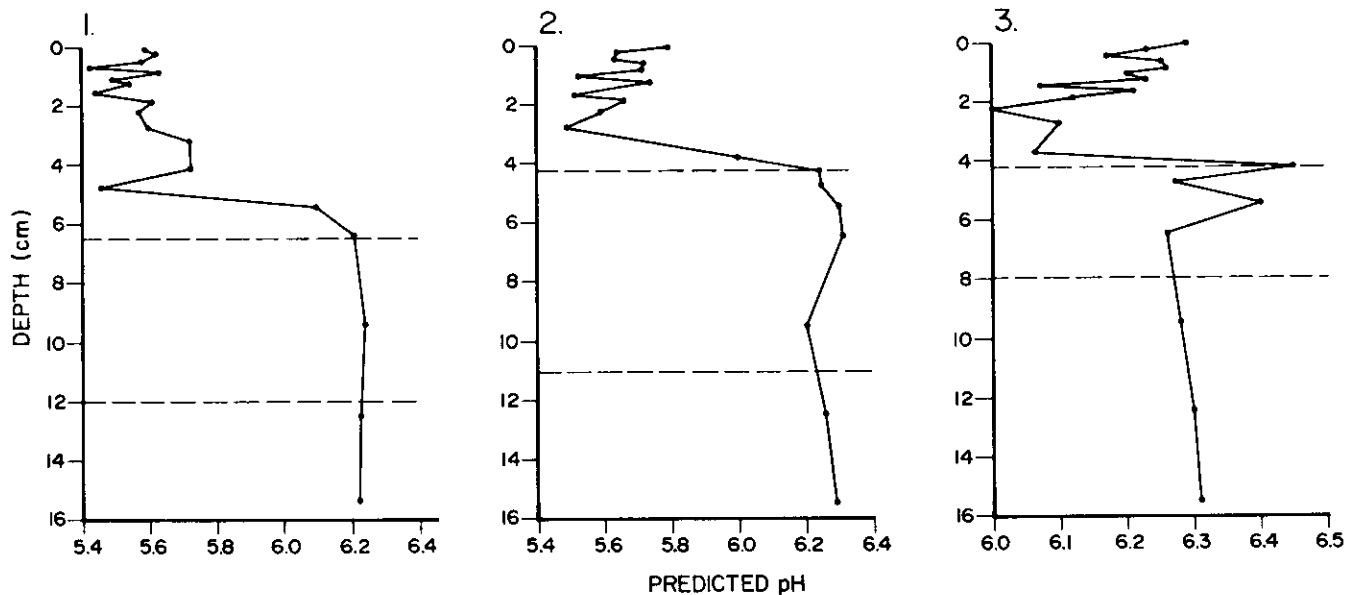


FIG. 3. Sediment core profiles of diatom-inferred pH, based on index B. Broken lines show the 3/4 pollen zone boundaries (lower) and the beginning of the decline in inferred pH (upper).

iment "focussing." Evans and Rigler (1980a, 1983) found that sedimentation rates varied directly with depth in three small Shield lakes in southern Ontario, with depth accounting for as much as 97% of the variation between sites. Accumulation rates were 2–3 times greater in the deep basins than at 10-m depths in these three lakes. Accumulation rates of 96–185 g·m⁻²·yr⁻¹ for recent sediments and 34–64 g·m⁻²·yr⁻¹

for premining sediments are likely close to the maxima for the three basins in Quirke Lake due to suspected relationships with depth. In comparison, Evans and Rigler (1980a, 1983) reported maxima of 226, 133, and 105 g·m⁻²·yr⁻¹ in three southern Ontario Shield lakes. Whole-lake averages for these three lakes were 45–78 g·m⁻²·yr⁻¹. In unproductive Toolik Lake, Alaska, Cornwell (1985) measured sedimentation rates of

12–73 $\text{g} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$. In unproductive Char Lake in the Canadian arctic, a whole-lake sedimentation rate of 31 $\text{g} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$ was estimated (de March 1978). G. J. Brunskill (pers. comm.) estimated average sedimentation rates of 22–56 $\text{g} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$ in four small arctic lakes in the eastern Keewatin District of Canada. These other data suggest that Quirke Lake has exhibited postmining sedimentation rates comparable with those reported for the other Shield lakes and premining sedimentation rates comparable with those found in unproductive arctic lakes. The sedimentation rates reported here in the deep basins of Quirke Lake are representative of potential areas that would be considered for deep lake disposal of tailings. Because tailings disposal would substantially alter the bathymetry of the lake by infilling of the deeper portions of the basin, then these sedimentation rates appear to represent upper limits for the rate of tailings burial following emplacement.

Dark grey to black laminations, separated by lighter reddish grey layers, were observed in the upper 8–12 cm of cores from Stations 1 and 3. No laminations were observed in cores from Station 2. Laminae were variable in thickness, but averaged about 1.4 mm at Station 1 and 0.5 mm at Station 3. If these laminae represent varves, as observed in other Sudbury-area lakes (Huhn 1974), then the inferred sedimentation rates (i.e. 1.4 and 0.5 $\text{mm} \cdot \text{yr}^{-1}$) are within the range of rates estimated for pre- and post-mining sediments at these stations (Table 1). The presence of these laminations provides evidence that the sediments are relatively undisturbed by processes such as bioturbation.

Core Profiles

Profiles of ^{238}U and ^{232}Th decay chain radionuclides all show enrichment in the surficial 5–7 cm (Fig. 4). The degree of enrichment relative to radionuclide activities in the deep sediments (≥ 9 cm) is 1–3 orders of magnitude for all radionuclides. Many profiles show diminishing concentrations between the maxima and more surficial sections, apparently demonstrating the effects of recent improvements in wastewater management. The zone of radionuclide enrichment in all cores extends to depths that coincide with or predate the beginning of mining and milling in the Quirke basin. The apparent low level of sediment mixing at Stations 1 and 3, as evidenced by sediment laminations, suggests that the downcore migration of some radionuclides at these stations may be attributed to diffusion. The apparent absence of laminations at Station 2 suggests that sediment mixing may contribute to this downcore migration at this location.

U concentrations of $>1000 \mu\text{g} \cdot \text{g}^{-1}$ in the surficial core sections of Quirke Lake (Fig. 4) are high relative to sediment U concentrations reported elsewhere. NRCC (1983) reported 1.45–24.7 $\mu\text{g} \text{U} \cdot \text{g}^{-1}$ in stream and lake sediments from across Canada. Brunskill and Wilkinson (1987) found 5–6 $\mu\text{g} \text{U} \cdot \text{g}^{-1}$ in suspended sediments from Lake 239, and G. J. Brunskill (pers. commun.) found approximately 2–6 $\mu\text{g} \text{U} \cdot \text{g}^{-1}$ in bottom sediments from eastern Canadian arctic lakes. In a geochemical survey of 469 lakes in northern Saskatchewan, Coker and Dunn (1983) reported background U concentrations of up to 228 $\mu\text{g} \cdot \text{g}^{-1}$ in sediments in zones of uranium mineralization, although the geometric mean concentration observed was only 2 $\mu\text{g} \cdot \text{g}^{-1}$. In Port Hope Harbour, Lake Ontario, U concentrations in surficial sediments also exceed 1000 $\mu\text{g} \cdot \text{g}^{-1}$ in some locations, due to inputs of residues from uranium and radium refining (Hart et al. 1986). Background concentrations

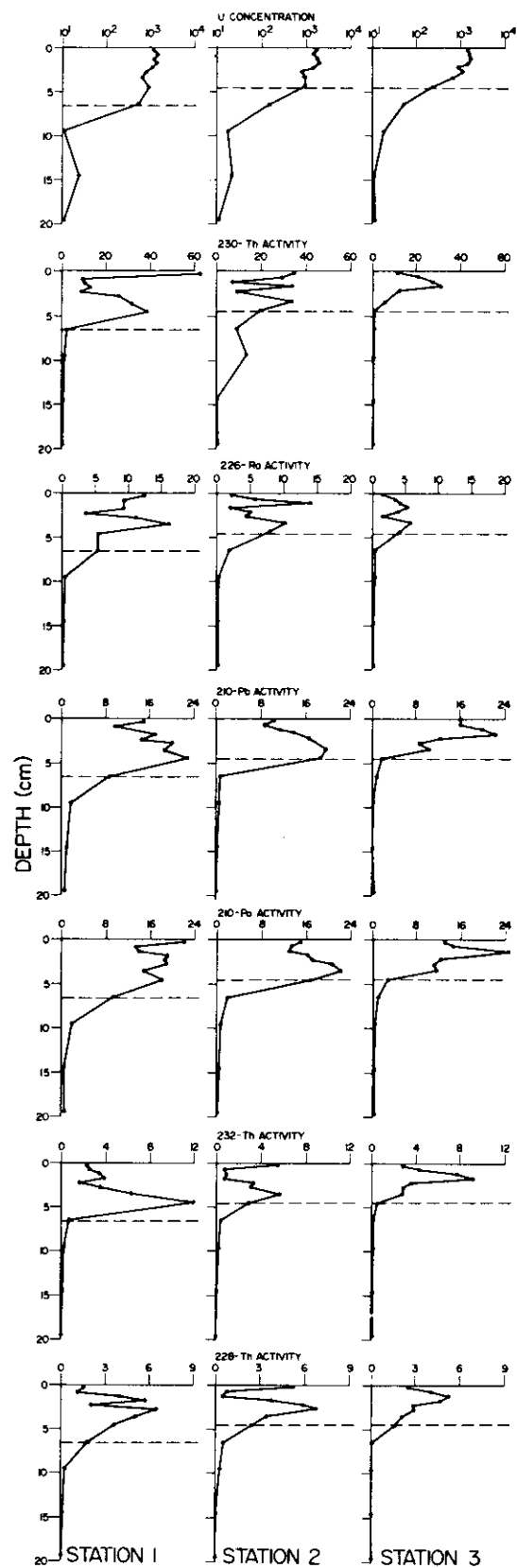


FIG. 4. Sediment core profiles of ^{238}U and ^{232}Th decay chain radionuclides at Stations 1, 2, and 3. U concentrations are in $\mu\text{g} \cdot \text{g}^{-1}$. Radionuclide activities are in $\text{Bq} \cdot \text{g}^{-1}$. Broken lines correspond to the beginning of uranium mining and milling (1957), estimated from the diatom-inferred pH decline (Fig. 3).

TABLE 2. Activity ratios in the surficial sediments of Quirke Lake and Serpent Harbour based on mean activities in the surficial 2.5 cm of Quirke Lake cores (this study) and on the mean activities in the surficial 2 cm of sediments from 18 sediment samples collected in Serpent Harbour (Hart and McKee 1985).

	$^{230}\text{Th}/^{238}\text{U}$	$^{226}\text{Ra}/^{230}\text{Th}$	$^{210}\text{Pb}/^{226}\text{Ra}$	$^{228}\text{Th}/^{232}\text{Th}$
Station 1	1.39	0.41	1.6	1.1
Station 2	1.14	0.26	2.0	1.4
Station 3	1.22	0.17	4.9	0.71
Serpent Harbour	0.33	14	0.76	4.3

of 11–26 $\mu\text{g U}\cdot\text{g}^{-1}$ in the deepest two sections of each Quirke Lake core tend toward the high end of the range of U concentrations typically found in lake sediments, apparently reflecting uranium mineralization in the Quirke Lake watershed. The maximum U concentrations in the Quirke Lake cores are equivalent to about 12–15 $\text{Bq}\cdot\text{g}^{-1}$ on a ^{238}U activity basis. These activities are somewhat less than the maximum activities of ^{230}Th , ^{210}Pb , and ^{210}Po and may reflect not only the greater mobility of U in the aquatic environment, but also the preferential extraction of U in the milling operation. Concentration profiles (Fig. 4) show that the U-enriched layer extends for several centimetres below the 1957 level. This can be attributed to the relatively high mobility of U in the environment (Dyck 1978). Diffusion of uranyl ions downward through the sediment porewater from an oxidizing surficial sediment layer and precipitation as UO_2 in a more reducing, deeper sediment layer may explain this phenomenon.

The activity of ^{230}Th , the first long-lived decay product of ^{234}U , ranges between 10 and 35 $\text{Bq}\cdot\text{g}^{-1}$ in surficial sediments of Quirke Lake (Fig. 4). While ^{230}Th activities in the surficial sediments of Quirke Lake are high, Hart et al. (1986) reported even higher activities (up to 77 $\text{Bq}\cdot\text{g}^{-1}$) in contaminated sediments of Port Hope Harbour. Background ^{230}Th activities of ≤ 0.05 to 0.5 $\text{Bq}\cdot\text{g}^{-1}$ in the deepest two sections of each Quirke Lake core are high relative to 0.014–0.016 $\text{Bq}\cdot\text{g}^{-1}$ in suspended sediment in Lake 239 (Brunskill and Wilkinson 1987) and 0.014–0.048 $\text{Bq}\cdot\text{g}^{-1}$ in bottom sediment from eastern Canadian arctic lakes (G. J. Brunskill, pers. comm.). Maximum ^{230}Th activities exceed the maximum activities of other ^{238}U decay chain radionuclides (Fig. 4), possibly reflecting the very low mobility of Th in natural waters (Dyck 1978; Langmuir and Herman 1980). Profiles of ^{230}Th show relatively little downcore migration below the 1957 level at Stations 1 and 3, but show substantial enrichment below this level at Station 2 (Fig. 4). This observation may reflect some contamination of the lower profile sediments by the enriched upper layer during the coring process, or may be attributed to sediment mixing at this location, since sediment laminations were not observed at Station 2. Diffusion of Th is unlikely, particularly since there is little evidence for downcore diffusion at the other two stations.

The $^{230}\text{Th}/^{238}\text{U}$ activity ratios, calculated as means for the top five core intervals, range from 1.14 at Station 2 to 1.39 at Station 1 in Quirke Lake sediments (Table 2). In the surficial sediments of Serpent Harbour, a waterbody at the mouth of the Serpent River watershed, Hart and McKee (1985) observed an average $^{230}\text{Th}/^{238}\text{U}$ activity ratio of 0.33 for the surficial sediments (Table 2). Th shows a greater affinity for sediment than does U, with a typical particulate–water distribution coefficient of >6500 for Th and 4000 for U in freshwaters (Santschi 1984). Based on the difference in $^{230}\text{Th}/^{238}\text{U}$ ratios in the upper

and lower Serpent watershed and on the greater affinity of Th for the sediment phase, it appears that U is transported downstream much more readily than Th.

The activity of ^{226}Ra , the decay daughter of ^{230}Th , ranges from 2 to 17 $\text{Bq}\cdot\text{g}^{-1}$ in the surficial enriched sediment of Quirke Lake cores, with activities in most surficial core sections of <10 $\text{Bq}\cdot\text{g}^{-1}$ (Fig. 3). ^{226}Ra activities of up to 297 $\text{Bq}\cdot\text{g}^{-1}$ have been observed in the sediments of Port Hope Harbour (Hart et al. 1986). ^{226}Ra activities of 0.032–0.047 $\text{Bq}\cdot\text{g}^{-1}$ were reported in suspended sediment of Lake 239 (Brunskill and Wilkinson 1987) and 0.008–0.023 $\text{Bq}\cdot\text{g}^{-1}$ in bottom sediments of eastern Canadian arctic lakes (G. J. Brunskill, pers. comm.); these are below background activities of 0.06–0.28 $\text{Bq}\cdot\text{g}^{-1}$ found in the deepest sections of the Quirke Lake cores. ^{226}Ra activities in the enriched sections of the Quirke Lake cores tend to be slightly lower than the activities of ^{238}U and ^{230}Th , with $^{226}\text{Ra}/^{230}\text{Th}$ ratios of 0.17–0.41 for the Quirke Lake cores (Table 2). In contrast, we found ^{226}Ra activities in Serpent Harbour sediments to be about an order of magnitude higher than activities of ^{230}Th (Table 2), indicating that ^{226}Ra is transported downstream more readily than its radioactive parent through the watershed. This is attributed to the lower sediment–water partition coefficient of Ra (typically 200–500; Kirchmann et al. 1966; Nishiwaki et al. 1972; Rancon 1973; Serne et al. 1977) relative to Th.

^{210}Pb , the decay product of ^{226}Ra , occurs in the surficial enriched sediments at activities of 10–23 $\text{Bq}\cdot\text{g}^{-1}$ (Fig. 4). In comparison, ^{210}Pb activities of up to 682 $\text{Bq}\cdot\text{g}^{-1}$ were reported in the contaminated surficial sediments of Port Hope Harbour (Hart et al. 1986). The excess of ^{210}Pb relative to ^{226}Ra of up to several Becquerels per gram in the surficial sediments of Quirke Lake results from differences in loading rates of the two radionuclides from mining and milling and the generally greater affinity of Pb for sediments (Snodgrass et al. 1983, 1986; Santschi 1984). The total ^{210}Pb activity from fallout and decay of in situ ^{226}Ra is typically ~ 0.2 –0.4 $\text{Bq}\cdot\text{g}^{-1}$ in Shield lake sediments (Evans and Rigler 1980a, 1983; Brunskill and Wilkinson 1987); thus, the contribution of fallout ^{210}Pb to substantially greater excesses in Quirke Lake sediments is relatively minor. ^{226}Ra and ^{210}Pb are in approximate equilibrium in the bottom sections of the cores, reflecting decay of excess ^{210}Pb from atmospheric fallout at the time of deposition to activities supported by decay of ^{226}Ra . Mean $^{210}\text{Pb}/^{226}\text{Ra}$ activity ratios of 1.6–4.9 in the surficial sediments of Quirke Lake and 0.76 in Serpent Harbour (Table 2) further demonstrate that ^{226}Ra is more mobile than ^{210}Pb and is more readily transported downstream.

Within stations, sedimentation rates in pre- and post-mining sediments are within the same order of magnitude (Table 1), while ^{210}Pb activities in each core range over three orders of magnitude (Fig. 4). Thus, the rate of ^{210}Pb supply has changed

markedly over time, and ^{210}Pb profiles cannot be used to estimate sedimentation rates using the "constant rate of supply" or "constant initial concentration" models (Appleby and Oldfield 1983).

^{210}Po , the granddaughter of ^{210}Pb , occurs in the surficial enriched sediment of Quirke Lake cores at activities similar to those of its grandparent (Fig. 4). A slight excess of ^{210}Po relative to ^{210}Pb is suggested in Station 1 and 2 profiles. This can be attributed to analytical error only, since ^{210}Po is supported in the sediment by decay of ^{210}Pb , with secular equilibrium enforced by the short half-life of ^{210}Po . Activities of ^{210}Po in Quirke Lake sediments are substantially lower than the maximum ^{210}Po activity reported in Port Hope Harbour sediments of $739 \text{ Bq} \cdot \text{g}^{-1}$ (Hart et al. 1986).

Activities of ^{232}Th and ^{228}Th of the ^{232}Th decay chain are $0.5-12 \text{ Bq} \cdot \text{g}^{-1}$ in the enriched layers of the Quirke Lake cores and $\leq 0.1 \text{ Bq} \cdot \text{g}^{-1}$ at the bottom of the cores (Fig. 4), with $^{228}\text{Th}/^{232}\text{Th}$ ratios ranging from 0.71 to 1.4 (Table 2). These activities are high relative to $0.025 \text{ Bq} \cdot \text{g}^{-1}$ for ^{232}Th typical of world soils (UNSCEAR 1982), $0.018-0.025 \text{ Bq} \cdot \text{g}^{-1}$ in suspended sediment from streams flowing into Lake 239 (Brunskill and Wilkinson 1987), $0.044-0.125 \text{ Bq} \cdot \text{g}^{-1}$ in bottom sediments from eastern Canadian arctic lakes (G. J. Brunskill, pers. comm.), and $0.014-5.2 \text{ Bq} \cdot \text{g}^{-1}$ in the most contaminated sediments of Port Hope Harbour (Hart et al. 1986). In Serpent Harbour sediments, activities of these radionuclides are 1-2 orders of magnitude lower than in Quirke Lake (Hart and McKee, 1985), suggesting that most of the loading of these radionuclides is sequestered in the sediments in the upper Serpent River system. A $^{228}\text{Th}/^{232}\text{Th}$ ratio of 4.3 in Serpent Harbour sediments is much greater than corresponding activity ratios of 0.71-1.4 in Quirke Lake (Table 2). ^{228}Ra is a relatively short-lived product (half-life of 6.7 yr) formed in the decay of ^{232}Th to ^{228}Th . A greater $^{228}\text{Th}/^{232}\text{Th}$ ratio in downstream sediments is apparently the result of more ^{228}Ra being transported than ^{232}Th from tailings areas and through downstream waters. This hypothesis is supported by limited (unpublished) data showing an excess of ^{228}Ra of >2 orders of magnitude relative to ^{232}Th in Quirke Lake water. ^{228}Th forms relatively rapidly (half-life of 1.9 yr) due to ingrowth from this displaced ^{228}Ra and is thereby transported to the sediments at activities exceeding those of ^{232}Th . This phenomenon should also result in $^{228}\text{Th}/^{232}\text{Th}$ ratios of >1 in Quirke Lake sediments, although the isotopic ratios observed ranged from 0.71 to 1.4 (Table 2) and do not show a definite ^{228}Th excess. The greater mobility of ^{228}Ra has also been reported to result in an excess of ^{228}Th relative to ^{232}Th in oceanic bottom waters (Nozaki et al. 1981).

The net sediment loadings at each of the three Quirke Lake stations may be estimated by multiplying the sediment radionuclide activities in the surficial sections of the cores (Becquerels per gram or micrograms per gram) by the postmining sedimentation rates (grams per square metre per year). These estimates are provided in Table 3 using mean activities or concentrations in the surficial five sections (2.5 cm) of the radionuclide profiles (Fig. 4). ^{210}Po and ^{228}Th are short-lived (half-lives of 138 d and 1.9 yr, respectively) relative to the period of sediment deposition represented by the surficial 2.5 cm. Thus, the activities of these radionuclides in the surficial layer are largely controlled by the activities of their long-lived grandparents, ^{210}Pb and ^{232}Th respectively. Therefore, "accumulation" rates for ^{210}Po and ^{228}Th (Table 3) include the effects of radioactive ingrowth in the sediments. Premining

TABLE 3. Net annual radionuclide accumulation rates for the postmining period (and premining period) at Stations 1, 2, and 3, Quirke Lake, based on estimated mass accumulation rates for postmining sediments (Table 1) and on mean concentrations or activities in the surficial 2.5 cm of sediment cores (Fig. 4). Values in parentheses are based on mass accumulation rates for premining sediments (Table 1) and on mean concentrations or activities in the bottom two core sections (Fig. 4). Units are $\text{mg} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$ for U and $\text{Bq} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$ for others.

Radionuclide	Accumulation rate		
	1	2	3
U	230 (1.0)	160 (1.1)	130 (0.4)
^{230}Th	4000 (19)	2200 (25)	2000 (≤ 2)
^{226}Ra	1600 (11)	570 (9.3)	340 (2)
^{210}Pb	2600	1200	1700
^{210}Po	3200	1400	1600
^{232}Th	500 (5)	210 (2)	530 (≤ 2)
^{228}Th	540	310	370

period sediment loadings cannot be calculated from radionuclide levels in sediments above the Zone 3/Zone 4 pollen boundary, due to the variable degree of downcore contamination. However, premining loadings may be estimated based on levels of long half-life radionuclides in core sections from below the Zone 3/Zone 4 boundary, assuming that sedimentation rates estimated for the 1835-1957 period also apply to these older deposits (Table 3). Estimated postmining sediment loadings in Quirke Lake are about 2 orders of magnitude greater than those estimated for the premining period and >1 to 3 orders of magnitude greater than those found in uncontaminated arctic lakes (G. J. Brunskill, pers. comm.).

The radiological consequences of radionuclide releases to aquatic systems from uranium mining and milling are the focus of concern of regulatory agencies and the general public. Sedimentation is recognized as a mechanism controlling the downstream transport of radionuclides (e.g. Snodgrass et al. 1983, 1986; Santschi 1984). Estimates of net sediment loadings of radionuclides, as provided herein, are required in the construction of environmental models necessary for the prediction of radionuclide transport and biological impact.

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