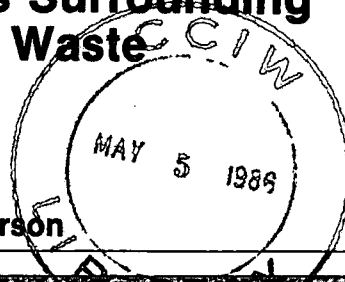**I✶** Environment Environnement
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Retention of ^{137}Cs and ^{90}Sr by Mineral Sorbents Surrounding Vitrified Nuclear Waste

K.E. Lyon and R.J. Patterson

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Cover photograph—

Split core RA5, glass block burial site.

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Abstract

In June 1960, twenty-five 14-cm diameter hemi-spheres of vitrified nuclear waste (glass blocks) were buried in a shallow sand aquifer in the lower Perch Lake Basin, Chalk River Nuclear Laboratories, Ontario, Canada. Almost all leaching of the glass blocks occurred during the first 18 months of emplacement, and consequently the burial experiment approximated a single-injection source of ^{137}Cs and ^{90}Sr. After about 11 years, most of the ^{90}Sr that was released had been advected and dispersed from the site, whereas the bulk of the ^{137}Cs had travelled no more than 0.3 m from the blocks.

Three piezometers were installed and five cores of sediment were collected at the glass block site in 1978. Mineralogically pure segregates of sand grains were prepared by hand and characterized using binocular, petrographic and scanning electron microscopy, X-ray diffraction, and energy-dispersive X-ray spectroscopy. A technique also was developed to measure very low levels of ^{137}Cs and ^{90}Sr by counting small (10–100 mg) sediment samples directly with a thin window, flow proportional counter.

Results show that sand-sized grains of altered biotite (biotite-vermiculite), biotite, muscovite, hornblende and sericitized feldspar are able to retain ^{137}Cs over a period of 18 years, and that grains of biotite-vermiculite, biotite and hematite-bearing feldspar (largely microcline) can retain low levels of ^{90}Sr over the same period of time. The amounts of ^{137}Cs and ^{90}Sr sorbed by the mineral grains ranged from undetectable levels to approximately 380 and 35 becquerels/gram sediment, respectively. The complete series of mineral segregates can be ranked on a weight basis according to the amounts of retained ^{137}Cs and ^{90}Sr. For ^{137}Cs the ranking is:

biotite-vermiculite >
biotite >
opaque or altered feldspar >
muscovite >
hornblende $\geq$
translucent feldspar $\geq$
quartz

and for ^{90}Sr:

biotite-vermiculite $\geq$
biotite and hematite-bearing feldspar >
all other mineral segregates (quartz, white feldspar, hornblende and muscovite).

Ground-water quality data indicate that none of the retention is the result of the precipitation of strontium or cesium minerals. Results for the mineral segregates, however, suggest that iron oxides play a significant role in the retention of ^{90}Sr, whereas ^{137}Cs appears to be retained principally as an interlayer cation in layer silicates. Results of other studies and calculations incorporating ground-water ^{90}Sr concentration data and experimental K_d^{Sr} values for the minerals indicate that a spectrum of adsorption sites, ranging from readily reversible to very slowly reversible, exists on the sediment for ^{90}Sr.

Résumé

En juin 1960, 25 hémisphères de déchets nucléaires vitrifiés de 14 cm de diamètre (blocs de verre) ont été enfouis dans un aquifère sablonneux et peu profond de la partie inférieure du bassin du lac Perch, aux Laboratoires nucléaires de Chalk River, en Ontario. La lixiviation des blocs s'est presque toute produite durant les 18 premiers mois, ce qui fait que l'expérience équivalait à une injection ponctuelle de ^{137}Cs et de ^{90}Sr. Après environ 11 ans, la majeure partie du ^{90}Sr libéré s'était déplacée horizontalement et dispersée, tandis que presque tout le ^{137}Cs ne s'était déplacé que de 0.3 m des blocs.

En 1978, on a installé trois piézomètres et extrait cinq carottes des sédiments du lieu d'enfouissement. Des grains de sable purs du point de vue minéralogique et séparés par ségrégation ont été préparés à la main puis caractérisés à l'aide de microscopes binoculaires, polarisants et électroniques à balayage, par diffraction des rayons X et par spectroscopie à rayons X dispersive. On a également mis au point une technique pour mesurer de très petites quantités de ^{137}Cs et de ^{90}Sr en comptant de petits échantillons de sédiments (de 10 à 100 mg) au moyen d'un compteur proportionnel à fenêtre mince.

Les résultats montrent que des particules de biotite altérée (biotite-vermiculite), de biotite, de muscovite, de hornblende et de feldspath séricitisé, de la grosseur des grains de sable, sont capables de retenir le ^{137}Cs pendant une période de 18 ans et que des particules de biotite-vermiculite, de biotite et de feldspath contenant de l'hématite (microline en grande partie) peuvent retenir de faibles quantités de ^{90}Sr sur une même période. Les quantités de ^{137}Cs et de ^{90}Sr adsorbées par les particules minérales variaient de l'indécelable à environ 380 et 35

becquerels par gramme de sédiment, respectivement. Toute la série des minéraux séparés par ségrégation peut être classée pondéralement selon les quantités retenues de ^{137}Cs et de ^{90}Sr. Pour le ^{137}Cs :

biotite-vermiculite >
biotite >
feldspath opaque ou altéré >
muscovite >
hornblende $\geq$
feldspath translucide $\geq$
quartz

et pour le ^{90}Sr :

biotite-vermiculite $\geq$
biotite et feldspath contenant de l'hématite >
tous les autres minéraux (quartz, feldspath blanc, hornblende et muscovite).

Les données sur la qualité des eaux souterraines montrent que la rétention n'est aucunement le résultat de la précipitation de minéraux du strontium ou du césium. Les résultats concernant les ségrégations semblent toutefois montrer que les oxydes de fer jouent un rôle important dans la rétention du ^{90}Sr, tandis que le ^{137}Cs paraît surtout retenu comme cation entre les couches de silicates. Les résultats d'autres recherches et les calculs dans lesquels on tient compte des concentrations de ^{90}Sr dans l'eau souterraine et des valeurs expérimentales de K_d^{Sr} dans les minéraux montrent que l'adsorption de ^{90}Sr varie localement dans les sédiments, du rapidement réversible au très lentement réversible.

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INTRODUCTION

An understanding of the behaviour of radionuclides such as ^{90}Sr and ^{137}Cs in ground-water flow systems is essential for the safe management of nuclear wastes. Most chemical models that may be used to simulate the transport of radionuclides in ground-water flow systems incorporate a distribution coefficient (K_d) to evaluate retardation of the solutes by the aquifer materials (Wang and Anderson, 1982). The use of the distribution coefficient assumes that local equilibrium is readily attained. Field and laboratory evidence, however, indicates that all sorption reactions are not necessarily readily reversible and, particularly, that retention of cations such as ^{90}Sr and ^{137}Cs can occur.

The term "retention" is used in this paper to describe desorption which is relatively slow (i.e., time periods in excess of several days) and which, therefore, leads to non-equilibrium sorption. Retention of cations by mineral grains can occur as a result of chemical processes such as specific adsorption and phyllosilicate interlayer sorption, or as a consequence of physical processes such as diffusion into intergranular fractures.

Both Cs and Sr may be chemically retained by sediments, although the mechanism of retention is different for each element. The retention of Cs occurs as a result of the so-called fixation of trace amounts of Cs as an interlayer cation in 2:1 phyllosilicates such as illite and the micas that have an interplanar spacing of approximately 10 angstroms. Although vermiculite typically has an interplanar spacing of 14.4 angstroms (Deer et al., 1966), it is also a "Cs-fixer" because the vermiculite interlayer will collapse with K or Cs saturation. Most of the early research, summarized by Tamura (1964), on the sorption of ^{137}Cs by these and other phyllosilicates was conducted in laboratories with reference minerals. Analysis of mineral segregates obtained from contaminated sediments later confirmed the great selectivity of vermiculite and the 10-angstrom phyllosilicates for ^{137}Cs (Francis and Brinkley, 1976).

The retention of Sr by sediments has been noted to occur as a result of specific adsorption by oxides of Fe, Mn and Al (throughout this paper, the term "oxide" is used in a general sense and is meant to include hydrous and anhydrous oxide, hydroxide, and oxyhydroxide compounds). Tamura and Struxness (1963) and Tamura (1965) reported that alumina and hydrous Fe and Al oxides have a high sorptive selectivity for Sr with respect to Na. Later, Murray (1975) and Kinniburgh et al. (1975) demonstrated that hydrous Mn dioxide, hydrous Fe and Al oxides, and amorphous Al hydroxide specifically adsorb trace amounts of Ca and Sr at solution pH values above and below the points of zero charge through a proton-metal exchange reaction.

A long-term experiment carried out at the Chalk River Nuclear Laboratories (C.R.N.L.) (Fig. 1) provided an opportunity to study retention of ^{90}Sr and ^{137}Cs. In June 1960, twenty-five 14-cm diameter hemispheres of vitrified nuclear waste were buried below the water table in the lower Perch Lake Basin (Fig. 1). Subsequent monitoring of ground water and aquifer sediments by staff of Atomic Energy of Canada Limited indicated that most of the radionuclides leached from the hemispheres were released within 18 months after burial (Merritt, 1976). As a result of the rapid leaching, the release can be considered to have approximated a single-injection tracer experiment.

The principal objective of the present study, which was conducted 18 years after burial of the blocks, was to identify the major minerals which retain ^{137}Cs and ^{90}Sr in sediment near the glass blocks. To achieve this objective, it was necessary to develop a radioanalytical technique that would permit the simultaneous determination of low levels (below 380 Bq/g sediment) of ^{137}Cs and ^{90}Sr in small (10 to 20 mg) mineralogically pure samples that could be separated from whole sediments.

PREVIOUS WORK

Hydrogeology

Shallow sediments in the lower Perch Lake Basin are predominantly well-sorted fine sands that were deposited during a high stage of the Ottawa River between about
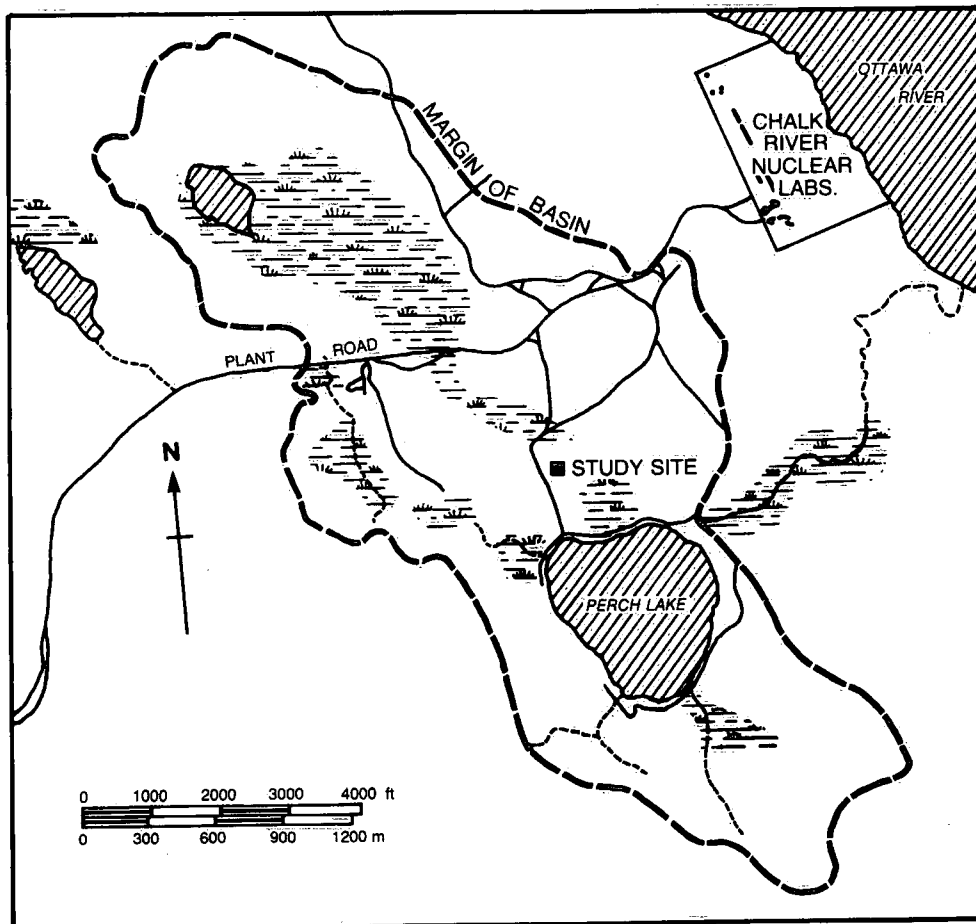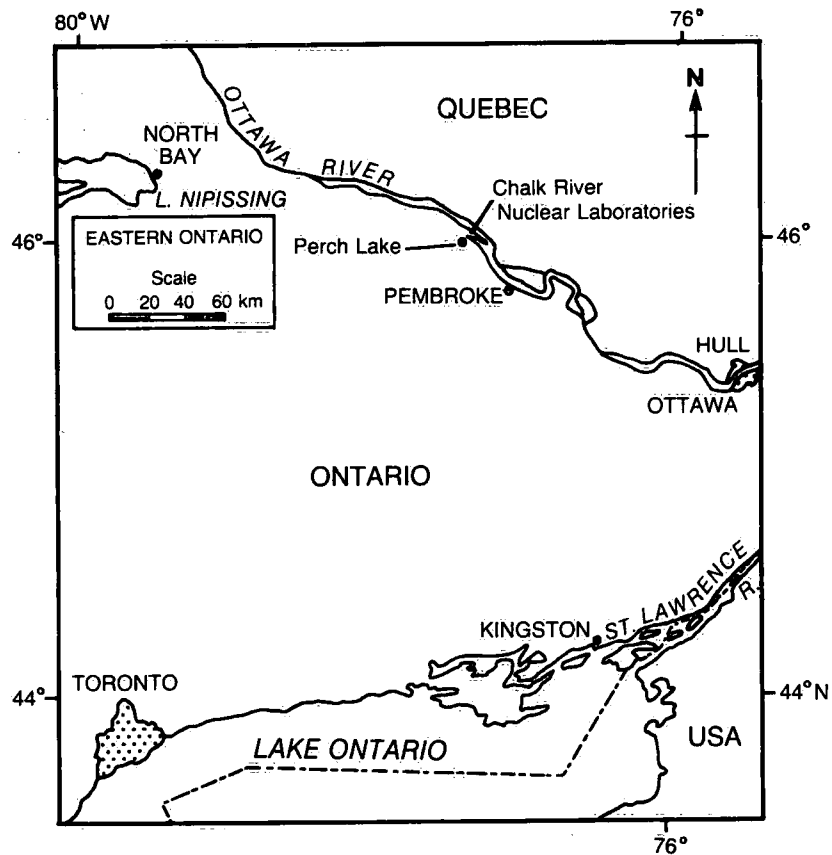
Figure 1.   Location of Chalk River Nuclear Laboratories and the study site in the Perch Lake Basin.

10 300 and 9800 years B.P. (Catto *et al.*, 1982). The mineralogy of the sands reflects their origin from underlying Precambrian Grenville Province bedrock, which in the area is composed largely of granitic gneiss and granite. The major minerals in the sediments are quartz and sericitized plagioclase (albite to andesine); the minor minerals are sericitized potassium feldspar, biotite, interstratified biotite-vermiculite, and hornblende (Jackson and Inch, 1980). The cation exchange capacity of the sediments is between 0.3 and 1.75 meq/100 g and the principal exchangeable cation is calcium (Jackson and Inch, 1980; Patterson and Spoel, 1981).

The depth to ground water varies seasonally, but is generally between 1 and 1.5 m. Horizontal hydraulic conductivities for the sands have been estimated by a variety of methods to range between 1×10^{-3} and 5×10^{-3} cm/s (Cherry *et al.*, 1975). Shallow ground waters flow at an average horizontal velocity of approximately 15–20 cm/day in a southeasterly direction (Merritt and Parsons, 1964; Pickens *et al.*, 1980).

Radionuclide Studies

The 25 radioactive glass blocks were buried at the study site in 1960 to investigate the leaching rates of irradiated uranium fuel reprocessing wastes which had been encapsulated in nepheline syenite glass (Merritt and Parsons, 1964). The blocks, each containing approximately 850 GBq of ^{137}Cs, 740 GBq of ^{90}Sr, 70 GBq of ^{144}Ce, and 7 GBq of ^{106}Ru, were placed in a two-dimensional, vertical grid pattern. The grid was oriented approximately perpendicular to the direction of ground-water flow in the depth interval 3.0 to 3.7 m below the ground surface (Figs. 2 and 3). Atomic Energy of Canada Limited staff have studied the leaching of the radioactive glass and the release of ^{137}Cs and ^{90}Sr to the ground-water flow system by regular monitoring of ground-water and aquifer sediments, direct examination of a hemisphere that was retrieved from the site in 1978, laboratory soil column and glass block leaching experiments, and mathematical modelling of the field monitoring results.

Average annual concentrations of ^{90}Sr in ground water 1.8 m downgradient from the blocks decreased from
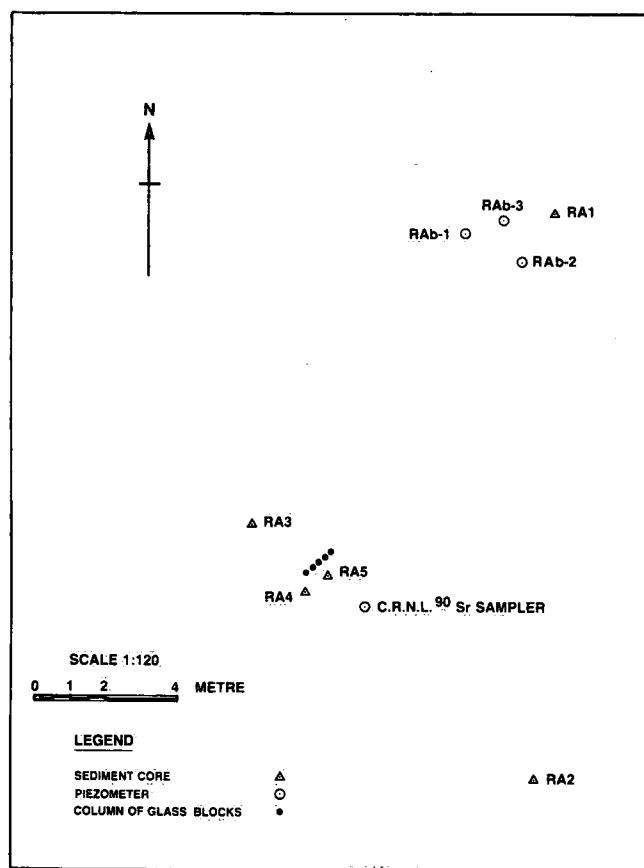

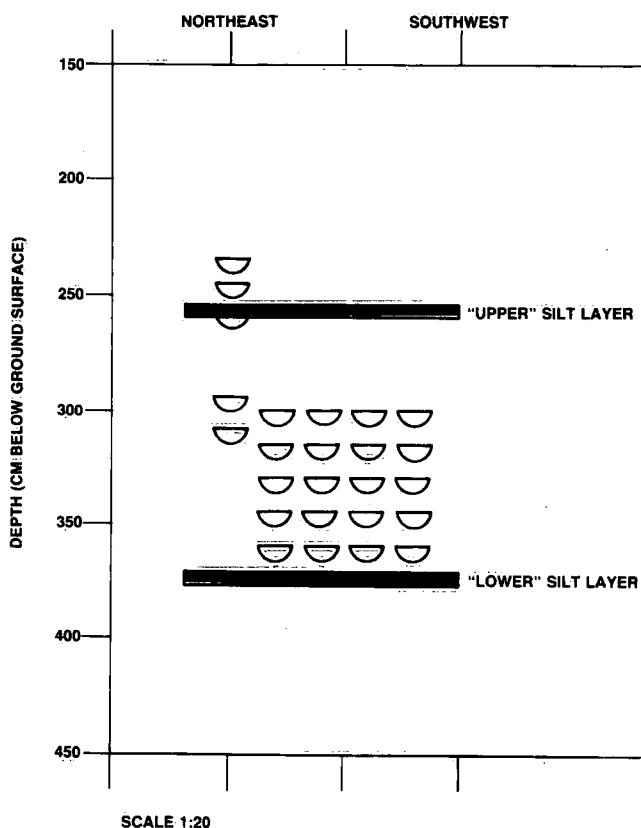Figure 2.   Location of the glass blocks, piezometers and cores at the study site.

Figure 3.   Vertical distribution of glass blocks (after Merritt and Parsons, 1964).

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4.8×10^3 Bq/L (1.3×10^{-1} μCi/L) in 1960 to 4.4 Bq/L (1.2×10^{-4} μCi/L) in 1973 and 1974 (Merritt, 1976). Based on this observation, Merritt estimated that 94% (52 MBq) of the ^{90}Sr leached from the glass was released within 18 months after burial. In 1963, and as shown in Figure 4, a ^{90}Sr contaminant plume could be defined to a distance of 11.9 m downgradient from the blocks (Merritt and Parsons, 1964). By 1971, however, because of transport and dispersion by ground-water flow, only very low levels of ^{90}Sr remained (W.F. Merritt, unpublished data).

Walton and Merritt (1979) compared the results of the 1963 soil survey (Fig. 4) with those predicted by a one-dimensional convective dispersive ground-water transport model, assuming a K_d^{Sr} of between 4.6 and 7 mL/g. The predicted results agreed within an order of magnitude with the measured ^{90}Sr soil concentrations, yet some discrep-ancies were noted. A better predicted fit to the field data was obtained later by Melnyk *et al.* (1984), who assumed that two sorption mechanisms were operating: an equilibrium ion exchange reaction and a disequilibrium oxyhydroxide sorption reaction. The half-life of the oxyhydroxide sorption reaction was estimated to be 2.0 ± 0.2 years.

Compared with ^{90}Sr, the transport of ^{137}Cs released at the site has been markedly different (Fig.4). Owing to strong sorption by the sediments, ^{137}Cs has never been detected in ground-water samples at the site (W.F. Merritt, unpub. data). From the results of a sediment survey conducted in 1979, Walton and Merritt (1979) concluded that more than 90% of the total amount of ^{137}Cs leached from the glass had travelled no farther than 0.3 m (similar results were obtained during the 1963 soil survey). However, a low level plume of ^{137}Cs was detected up to 7.5 m downgradient of the blocks. Soil column experiments have subsequently indicated that this transport occurs on fine grain particulate matter (largely between 0.2 and 1.0 μm in diameter) and that formation and release of the particulates is microbiologically mediated (Champ and Merritt, 1981).

METHODS

Ground-Water Samples

Three piezometers constructed from 3.8 cm I.D. ABS plastic pipe were installed and developed 11 m northeast of the vertical grid of glass blocks (Fig. 2). The water table at the site was 1.3 m below ground surface, and the screened intervals of the piezometers were placed at depths between 1.8 and 5.7 m.

Water level measurements were taken in August and September 1978 and in February 1979, and the piezometer waters were sampled in August 1978 with a portable peristaltic pump. Field E_H and pH electrode measurements were taken in an airtight, in-line flow cell after several piezometer volumes of water had been removed. Ground-water conductivity, temperature and dissolved oxygen were also determined in the field at the time of sampling.

Samples for total inorganic carbon were retained in the field in disposable syringes and then analyzed with an Oceanography International Total Carbon System. Chloride was measured with an ion-selective electrode and alkalinity levels were determined by acid titration. Sulphate was measured turbidimetrically by precipitation as $BaSO_4$.

Ground-water samples for the rest of the analyses were filtered through 0.45-μm membranes in the laboratory under N_2 (gas) pressure. Those samples to be used for major
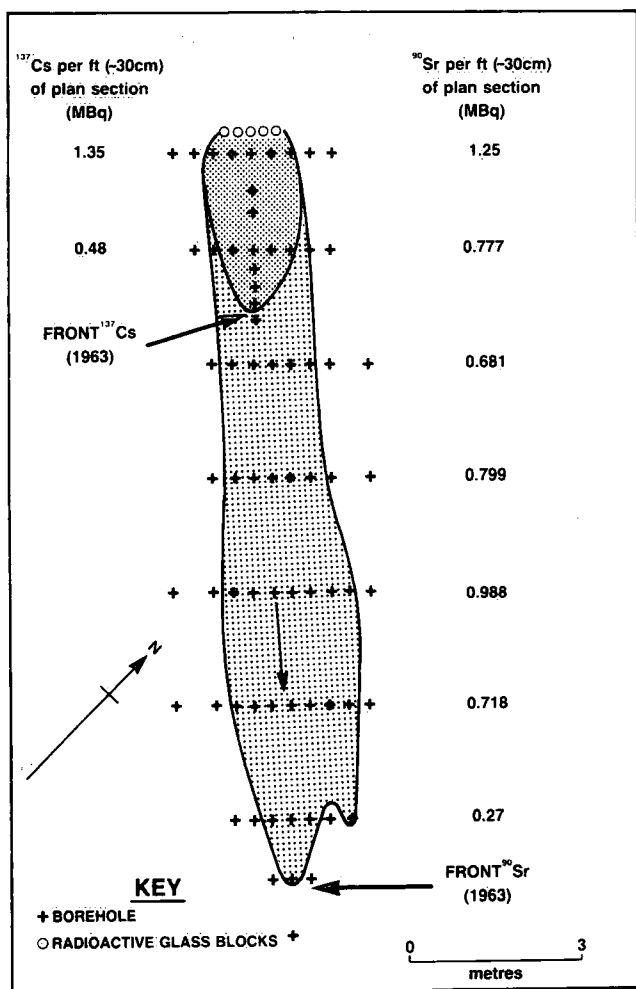

Figure 4. Plan view of ^{137}Cs and ^{90}Sr migration downgradient from the glass blocks in 1963 (after Merritt and Parsons, 1964).

cations were acidified with concentrated HCl prior to analysis by atomic absorption spectrophotometry. Concentrations of trace cations (Sr, Ba, Rb, Cs) were measured in non-acidified samples by a thermal neutron activation and gamma-ray spectroscopy technique (Merritt, 1971).

Sediment Samples and Characterization

Five sediment cores spanning the depth interval from 1.3 to 4.2 m were recovered from the second glass block site by procedures that have been described by Patterson et al. (1978). The cores, retained in 5.08-cm diameter thin-walled aluminum tubing, were numbered sequentially and prefixed with "RA", the designation for the closest C.R.N.L. piezometer nest.

Core RA1 was sectioned into 30-cm lengths and later used by Patterson and Spoel (1981) to determine K_d^{Sr} values. After gravity drainage of water, all other cores were sectioned longitudinally with a carbide-tipped saw blade. Sediment samples were then collected at selected intervals for freeze-drying and subsequent preparation for counting.

Each sediment sample was designated according to the core and depth of the top of the selected interval below ground level. For example, sample RA5:353 was obtained from the depth interval beginning at 353-cm depth below ground in core RA5. One additional sediment sample from the C.R.N.L. 1971 sediment survey was used in the study. This sample is designated 010:9 and was obtained from a depth of 272–276 cm at a location 0.3 m downgradient from the blocks.

Freeze-dried samples selected for grain size analysis were sieved for 10 min in a Ro-Tap shaker. The results of weighing the sieved fractions were plotted as cumulative curves on logarithmic probability paper, and moment measures were estimated graphically according to the methods of Folk and Ward (1957). Silt and clay laminae were sieved to a grain size of less than 0.0625 mm prior to sample preparation for counting.

The mineralogy of grains was determined under a reflecting binocular microscope on the basis of physical characteristics such as cleavage, fracture, colour and lustre. Samples of quartz, feldspar, biotite, biotite-vermiculite, muscovite and hornblende were segregated from the 0.42- to 0.50-mm grain size fraction with the use of fine jeweller's tweezers or a sharp needle.

Further characterization of the grains was carried out using thin-sections, scanning electron microscopy-energy dispersive X-ray spectrometry (SEM-EDS), and X-ray diffraction analysis. The SEM used at C.R.N.L. was a

Cambridge Stereoscan, which was coupled with a Tracor Northern EDS unit. The SEM samples were not pretreated except by freeze-drying and carbon-coating. A semi-quantitative log energy intensity scale was used in distinguishing elements heavier than neon in major, minor and trace amounts. The X-ray diffraction analysis was undertaken at Queen's University with a Picker diffractometer.

Sample Preparation for Counting

Three types of sediments were prepared for counting: whole sediments, grain size fractions and mineral segregates. Whole samples representing stratigraphic intervals and sieved silt and clay samples were taken directly by spatula from thoroughly shaken sample bottles and distributed as evenly as possible and to a thickness of less than 1 mm over individual pre-weighed, 3.0-cm diameter stainless steel counting disks. Sieved grain size fractions were taken from mechanically split fractions obtained with a Soiltest sediment splitter. The grains in the mineral segregates were distributed evenly in the centre of the disks. Following a second weighing, all samples were attached to the disk with flexible collodion.

The Gaussian standard deviation associated with sample weight measurements was calculated and used in all subsequent determinations of weight-normalized counting rates. Sandy sediment weights were generally between 40 and 100 mg, and silt and clay weights usually ranged from 20 to 35 mg. Weights of the mineral segregate samples generally ranged between 11 and 14 mg.

Sample Counting and Instrument Calibration

Sediment samples were counted in a Sharp low beta flow-proportional counter which uses P-10 gas (90% Ar and 10% CH_4). The counter contains two detectors with 100 $\mu g/cm^2$ windows and anti-coincidence shielding is provided by two connected guard counters. One source of low level extraneous radiation at C.R.N.L. is atmospheric ^{41}Ar, formed from ^{40}Ar exposed to the neutron flux in research nuclear reactors. Sediment samples were counted only when levels of atmospheric ^{41}Ar were judged to be insignificant or low as indicated by a NaI scintillation detector. When low levels of ^{41}Ar were indicated, the detector was surrounded by a continuous flow of N_2 gas to minimize the effect of ^{41}Ar on counting rates.

Samples were generally counted for 2 to 4 h, but always until at least 120 counts were collected. This length of time represented a compromise between obtaining adequate and consistent counting statistics, processing as many samples as possible, and minimizing the inconvenience of manual control of the counting periods.

Counting results are reported as gross, instrument background, corrected, and net counting rates (counts per minute, cpm). The corrected sample counting rate was calculated by subtracting the instrument background counting rate (determined with empty counting disks) from the gross counting rate. All corrected counting rates were then normalized to a sample weight of 100 mg, with the assumption that the detector counting efficiencies for natural terrestrial radionuclides within the sediment matrix are independent of sample weight. This assumption is considered to be valid for the low levels that were present (W.F. Merritt, personal communication). A net sample counting rate (cpm/100 mg) attributable to the presence of ^{137}Cs and ^{90}Sr was then calculated by subtracting the corrected counting rate of an uncontaminated, or blank, sediment sample. Natural terrestrial radionuclides which are present within sediments and which were represented by corrected blank counting rates include ^{40}K, ^{87}Rb, and members of the ^{238}U and ^{232}Th decay series.

To discriminate between the contribution of ^{137}Cs and ^{90}Sr to net counting rates, selected samples were counted with and without an aluminum absorber placed over the sample disk. The absorber, or filter, had a thickness corresponding to 225 mg/cm^2 which absorbs all beta particles with energies less than 0.63 MeV (Friedlander et al., 1964). Thus, the filter completely blocked the ^{90}Sr, ^{87}Rb and 0.51 MeV ^{137}Cs beta particles. Attenuated beta contributions to the filtered sample counting rate were possible from the decay of the ^{90}Sr daughter product, ^{90}Y, and the minor ^{137}Cs beta particle (maximum energy of 1.18 MeV), as well as from the decay of ^{40}K, the ^{238}U and ^{232}Th series of radionuclides, and ^{41}Ar. Trace gamma contributions to the filtered counting rate were possible from the decay of ^{137m}Ba, ^{40}K, the ^{238}Y and ^{232}Th decay series of radionuclides, and ^{41}Ar.

The ^{137}Cs and ^{90}Sr calibration standards were prepared from samples of uncontaminated fine sand that were spiked with known activities (53.7 ± 0.5 and 63.7 ± 0.6 Bq/g, respectively) of the two radionuclides. The counting efficiency of the radiation detectors was assumed to depend upon sample weight, and several subsplits of spiked and blank samples weighing between 10 and 100 mg were prepared. Net unfiltered and filtered counting rates of spiked samples were then used in constructing separate ^{137}Cs and ^{90}Sr calibration curves. Unfiltered detector efficiencies for both radionuclides were approximately 20% to 30%, depending upon sample weight. The filtered detector efficiencies for ^{137}Cs and ^{90}Sr were approximately 6% and 0.2% to 0.3%, respectively.

Counting Statistics and Calculation of Radioactivity

Counting and disintegration rates are listed with standard deviations attributable to counting statistics and the uncertainty in sample weights. Standard deviations of the gross counting rates were calculated as the square root of the total number of counts divided by the counting time, in accordance with the assumption of a Poisson distribution. Standard deviations of the instrument background counting rates, however, were estimated by the mean Gaussian sample standard deviation because of the occasional presence of minor levels of atmospheric ^{41}Ar.

Low counting rates were recorded for many of the samples, and it was necessary to establish critical levels for detection (L_c) of both corrected and net counting rates. Approximate values of the critical levels for detection were derived from the sample variances of counting rates, as described by Lyon (1981). A value of 0.10 was selected as the maximum acceptable probability of making an error of the first kind (deciding a substance is present when it is not).

Because it was not feasible to standardize counting times, it was not possible to calculate detection limits for counting rates as rigorously defined by radiochemists (for example, see Currie, 1968). Accordingly, an alternative procedure was developed that relies upon the calculation of the probability that a corrected or net counting rate (x) with sample standard deviation (S_x) exceeds the critical level for detection of the rate. The calculation was based on the expression

$$Z = \frac{L_c - x}{S_x}$$

where Z is the standard normal deviate. The cumulative normal frequency distribution greater than Z was then obtained from Snedecor and Cochran (1967) and is reported as the probability of detection.

For any sediment sample with statistically significant net unfiltered and filtered counting rates, the activity of ^{137}Cs and ^{90}Sr present in the sample can be calculated by solving two equations derived from several known relationships. The equations are described by Lyon (1981) and include terms related to detector counting efficiencies, unfiltered and filtered net counting rates of the samples, and the ratios of the unfiltered to filtered net counting rates of the calibration standards. Concentrations of both radionuclides are expressed as disintegrations per minute per 100 mg (dpm/100 mg) of dry sediment.

The approximate level of detection for both radionuclides was 10 dpm/100 mg (2 Bq/g). By way of comparison, an activity of 10 dpm/100 mg is equivalent to 5.2 × 10^{-13} g ^{137}Cs per gram dry sediment and 3.2 × 10^{-13} g ^{90}Sr per gram dry sediment.

The single greatest constraint on the simultaneous determination of ^{137}Cs and ^{90}Sr in sediments using the low beta flow proportional counter is the very low counting rates obtained when the aluminum absorber is in place. These low rates decreased the precision of the reported rates as well as the filtered ^{90}Sr calibration curve. For many of the samples, the probability was less than 90% that corrected and net filtered counting rates exceeded their critical levels for detection. If these counting rates were assumed to be zero, however, unwarranted bias possibly would have been given to the unfiltered counting rates. In this study, the criterion was adopted that any corrected counting rate which exceeded its critical level for detection (probability of detection greater than 50%) was used in further calculation of net counting rates. Counting rates that did not meet this criterion were reported as not detected and were assumed to be zero. Similarly, any net filtered counting rate with a probability of detection greater than 0.5 was used in further calculation of ^{137}Cs and ^{90}Sr activities.

Samples with undetected net filtered counting rates but with detected net unfiltered counting rates were assumed to contain only ^{137}Cs. However, ^{90}Sr could have been present in these samples, and a method was developed to indicate this possibility (Lyon, 1981). The calculated probability that ^{90}Sr activity exceeded zero dpm/100 mg is reported in the tables of data and in every case was less than 0.5.

Two whole sediment samples (RA5:353 and RA5:373), each weighing approximately 18 g, were analyzed with a Ge(Li) gamma spectrometer and then were mechanically split and analyzed with the low beta counter. Levels of ^{137}Cs determined with the low beta counter were greater than levels determined with the Ge(Li) spectrometer by approximately 20% to 55%. This apparent difference may be the result of systematic error in the calibration curves or of a mineralogic bias introduced by whole sediment sample preparation.

RESULTS

Ground-Water Levels and Chemistry

During August–September 1978 and February 1979, vertical hydraulic gradients were variable over the depth interval of 1.8 to 5.7 m at the site. The maximum measured gradient between the screened intervals of the three piezometers was 0.0106. This corresponds to an upward vertical

Table 1. Chemical Results for Ground Waters at Piezometer Nest RAb

Parameter*	Piezometer			
	RAb-1	RAb-2		RAb-3
Sample interval (cm below ground surface)	180–220	330–370		530–570
Date sampled	1978-08-01	1978-08-01	1978-08-24	1978-08-02
Temperature (°C)	–	–	8.6	8.0
Conductivity (μS/cm)	62	–	54	82
pH	5.4	–	5.5	6.4
E_H (V)	<0.3	0.44	0.55	0.27
H_2S (gas)	Present	Absent	Absent	Present
Dissolved oxygen†	0.2	1.2	–	0.1
Fe	0.3	0.1	<0.1	2.0
Mn	0.06	0.07	<0.05	0.06
Ca	6.5	5.5	7.5	12.5
Mg	1.6	1.8	2.2	3.6
Sr	3.5×10^{-2}	3.3×10^{-2}	3.7×10^{-2}	5.3×10^{-2}
Ba	1.6×10^{-2}	1.5×10^{-2}	1.8×10^{-2}	1.2×10^{-2}
Na	2.6	2.2	2.5	2.7
K	0.6	0.7	0.7	1.7
Rb	4.6×10^{-4}	1.8×10^{-3}	2.0×10^{-3}	1.4×10^{-3}
Cs	$<1 \times 10^{-6}$	5×10^{-6}	$<1 \times 10^{-6}$	$<1 \times 10^{-6}$
T.I.C.‡	11.0	11.3	11.5	12.6
Alkalinity (meq/L)	–	–	0.11	–
SO_4	–	–	18	–
Cl	7.8	5.6	–	21.6

* All units are in milligrams per litre unless noted, with the exception of pH.
† Measurement may include dissolved oxygen which diffused through the Tygon sampling tube.
‡ Total inorganic carbon.

velocity (assuming a sediment porosity of 0.38) of approximately 6 cm/day (Lyon, 1981), or only about one-third the value of average horizontal ground-water velocities at the site.

Analytical results for ground-water samples obtained at the site are given in Table 1. The chemistry of these waters is typical of shallow ground waters throughout the Perch Lake Basin.

Water sampled from the middle piezometer (RAb-2) had an ionic strength of 10^{-3} and calculated values of solubility indices indicated undersaturation with respect to calcite, strontianite and celestite. On the basis of this undersaturation and SEM-EDS studies which showed no evidence of mineral deposition on sediment grains, retention of ^{90}Sr cannot be accounted for by precipitation of solid phases. Similarly, Cs losses cannot be ascribed to mineral formation.

Concentrations of dissolved iron in water from the middle piezometer were less than or equal to 0.1 mg/L, and probably reflect the controlling influence of an iron oxide solid phase. No authigenic iron sulphide was observed by SEM-EDS analysis of site sediments.

Stratigraphy and Sedimentology

Soil at the site belongs to the orthic humo-ferric podzol subgroup (Canadian Soil Survey Classification) and the Bf horizon extends to a maximum depth of 1.5 m.

Using the terminology of Reineck and Singh (1973), sedimentary bedding in the cored interval 1.3 to 4.2 m below ground surface can be described as coarsely interlayered. The bedding consists of lamina sets (ranging in thickness to 50 cm or more) of well-sorted, fine sand separated by thin (1 mm) laminae of silt and clay. Figure 5 illustrates the stratigraphy observed in core RA5.

Some core sections displayed very distinct, even laminations of sand. These laminations, generally less than 1 cm thick, are composed of finer grained, mafic (biotite and hornblende)-rich layers and coarser grained, felsic layers and possibly represent a littoral environment. Other core sections displayed less distinct, thicker (to 5 cm) laminations of sand in which variations in the silt content often served to define the laminations. The sizes of cross-laminations of sand that were observed are similar to those produced by migrating small current or wave ripples.

A massive, medium grain feldspar-rich layer of 2- to 11-cm thickness was found in cores RA3, RA4 and RA5. It was the only layer in the cores that contained pebbles.
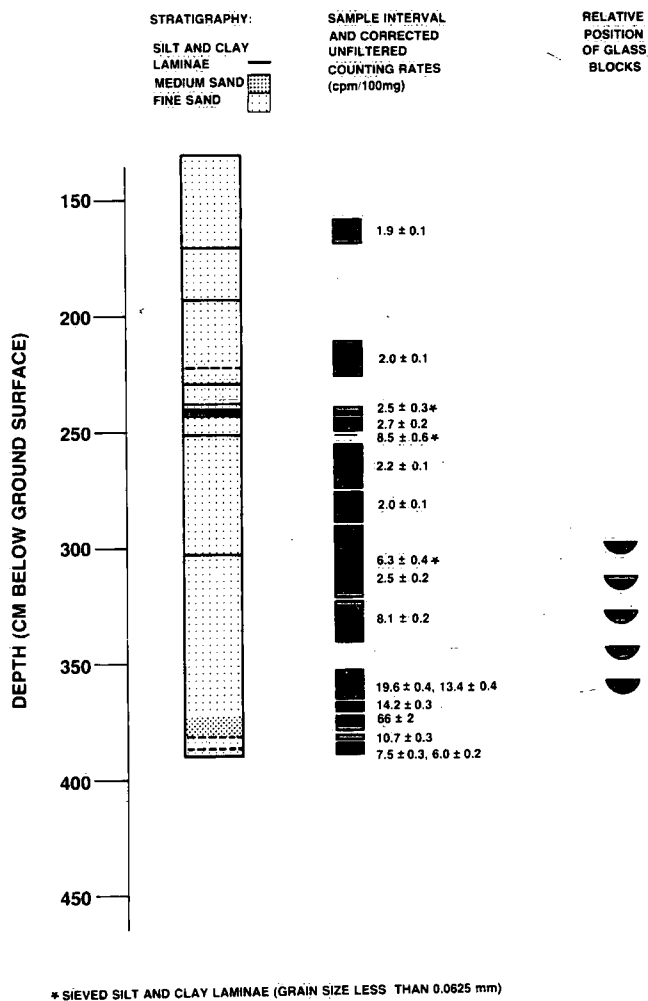
Figure 5. Sediment stratigraphy and distribution of radioactivity in core RA5.

Activity of Whole Sediment Samples

Whole sediment samples from the cores RA2, RA4 and RA5 were counted to determine the stratigraphic distribution of ^{137}Cs and ^{90}Sr contamination in the vicinity of the blocks. Samples from the 330- to 360-cm depth interval in core RA3, which was taken upgradient from the blocks, were used to establish background counting rates. The corrected unfiltered counting rate over 2 h for these background samples ranged between 1.3 ± 0.1 and 2.0 ± 0.2 cpm/100 mg. These rates were due solely to naturally occurring terrestrial radionuclides.

Counting rate results for sediments from the other cores indicate that contamination (26 cpm/100 mg) at RA2 was restricted to a 1-cm thick clay lamina at the 2.6-m depth. At RA4, low levels of contamination (up to 26 cpm/

100 mg) were indicated for silt laminae in the depth interval 3.2 to 4.1 m. The highest levels of contamination were encountered in core RA5 (Fig. 5) located only 40 cm down-gradient from the blocks. Contaminated sand samples were found at depths between 2.9 m and the bottom of the core, and the most active sample (66 cpm/100 mg) came from the medium grain felsic sand layer. Silt laminae in the depth interval 2.5 to 3.0 m below ground level show higher levels of contamination than adjacent fine sands. Since the highest levels of contamination were measured in RA5, subsequent detailed investigations were carried out on sediments from this core.

Sorption of ^{137}Cs and ^{90}Sr by Mineral Fractions

The distribution of sorbed ^{137}Cs and ^{90}Sr on the basis of mineralogy was determined for two contaminated sediment samples: RA5:353 (353- to 367-cm depth interval) and RA5:373 (373- to 380-cm depth interval). Segregates of quartz, several feldspars, biotite, biotite-vermiculite, muscovite and hornblende were prepared for counting from the 0.42- to 0.50-mm size fraction of each sediment sample. Blank counting rates attributable to natural radionuclides and ranging from not detected to 7 cpm/100 mg were obtained from uncontaminated mineral grains segregated from the same grain size fraction of RA3:346. The highest blank unfiltered counting rates (greater than 5 cpm/100 mg) were obtained with K-rich feldspar, muscovite and biotite and probably reflect the presence of ^{40}K. With alteration to vermiculite, biotite loses interlayer K, and this loss is reflected in the lower unfiltered counting rate (2.6 cpm/100 mg) of biotite-vermiculite.

The counting rates and levels of ^{137}Cs and ^{90}Sr sorbed by all the prepared mineral segregates are listed in Table 2.

Quartz

Quartz grains were identified under the reflecting microscope on the basis of colour, vitreous lustre and glacial surface textures. Most grains were angular to sub-angular with high sphericity (Powers, 1953). Some grains appeared to contain trace to minor surface patches of a red-orange iron oxide coating. In rare instances, an iron oxide-coated fracture plane through a grain could be observed. Petrographic analysis of a thin section prepared from a segregate of quartz grains showed that most grains were monocrystalline with undulatory extinction. The X-ray diffraction analyses indicated that the samples contained only trace or minor quantities of feldspar.

The SEM analysis showed that the quartz grains display well-developed glacial surface textures such as con-

choidal breakage patterns (Krinsley and Donahue, 1968; Krinsley and Doornkamp, 1973). None of the grains that was examined showed well-developed chemical dissolution or precipitation features. The quartz grains generally had abundant adhering particles, and the variable composition of this material as determined by X-ray intensity spectra suggested that most of the particles were glacial rock flour. Some iron-rich material was also detected on the grains and is likely iron oxide.

The counting rates and levels of radioactive contamination for the four quartz segregates are listed in Table 2. The net unfiltered counting rates that were obtained for these segregates were the lowest of all the mineral segregates that were prepared, with the single exception of one feldspar segregate. The highest net unfiltered counting rate (5.4 cpm/100 mg) and the only probable occurrence of ^{90}Sr (15 ± 10 dpm/100 mg) were associated with sample 373-Q3. The surfaces of all grains in this segregate displayed trace to minor amounts of a red-orange coating, which presumably was iron oxide.

Hornblende

Hornblende grains were segregated on the basis of their characteristic cleavage (producing surface angles of 56° and 124°), vitreous lustre and black colour. Trace amounts of brown silt and clay were visible on most hornblende grains observed under the reflecting microscope. In the grain size fraction examined, hornblende comprised less than 1% of the grain population.

Only one contaminated hornblende segregate was prepared, and it contained approximately 100 dpm ^{137}Cs/100 mg (Table 2). Evans (1954) has reported previously that hornblende segregates obtained from lower Perch Lake Basin sediments retain Cs. The mechanism of retention has yet to be determined, but may be related to incongruent mineral dissolution.

Feldspar

Characteristic cleavage, vitreous to pearly lustre, colour, and uneven to conchoidal fracture served as the main criteria in identifying feldspar. The segregated grains were translucent to opaque, sub-angular to subrounded and high in sphericity (Powers, 1953). In most cases, grains were segregated according to colour into two major groups: pink plus orange; and white. There is some feldspar in the sediments that resembles quartz but it was not possible to segregate it effectively using the reflecting microscope. About 20% of the segregated feldspar grains showed trace surface patches of iron oxide or silt-clay.

Table 2. Sample RA5:353 and RA5:373 and 010:9 (from 1971 Survey) Counting Rates and Radioactive Contamination of Segregated Mineral Grains

Sample number*	Type of count†	Counting rate (counts ÷ min)	Weight (mg)	Corrected counting rate (cpm/100 mg)	Probability of detection‡	Net counting rate (cpm/100 mg)	Probability of detection‡	^{137}Cs (dpm/100 mg)§	^{90}Sr (dpm/100 mg)§
353-Q1	UF	750 ÷ 788	12.66	2.13 ± 0.35	1.00	N.D.	0.12	N.D.	N.D.
	F	99 ÷ 121	12.66	N.D.	0.00	N.D.	0.00		
373-Q1	UF	1118 ÷ 946	10.92	4.57 ± 0.41	1.00	2.79 ± 0.82	0.97	9.4 ± 2.7	N.D.
	F	167 ÷ 161	10.92	N.D.	0.49	N.D.	0.49	(P = 1.00)	(P = 0.33)
373-Q2	UF	168 ÷ 126	14.32	4.54 ± 0.74	1.00	2.76 ± 1.02	0.83	—	—
373-Q3	UF	230 ÷ 131	15.03	7.14 ± 0.77	1.00	5.36 ± 1.05	1.00	3 ± 12	15 ± 10
	F	148 ÷ 136	15.03	0.95 ± 0.61	0.70	0.95 ± 0.61	0.70	(P = 0.55)	(P = 0.83)
373-H1	UF	640 ÷ 139	13.54	29.0 ± 1.4	1.00	29.0 ± 1.4	1.00	99.9 ± 5.5	N.D.
	F	133 ÷ 145	13.54	N.D.	0.07	N.D.	0.00	(P = 1.00)	(P = 0.36)
353-F4 (translucent pink)	UF	365 ÷ 144	12.82	14.4 ± 1.1	1.00	9.40 ± 1.12	1.00	N.D.	29.7 ± 3.6
	F	149 ÷ 120	12.82	2.31 ± 0.81	0.97	2.31 ± 0.81	0.97	(P = 0.32)	(P = 1.00)
353-F6 (translucent pink)	UF	340 ÷ 156	11.75	12.7 ± 1.0	1.00	7.69 ± 1.10	1.00	26.0 ± 3.8	N.D.
	F	668 ÷ 647	11.75	N.D.	0.43	N.D.	0.43	(P = 1.00)	(P = 0.12)
353-K1 (Opaque orange)	UF	662 ÷ 169	9.77	33.1 ± 1.6	1.00	27.8 ± 1.8	1.00	58.3 ± 9.1	32.3 ± 6.3
	F	1409 ÷ 1206	9.77	2.28 ± 0.38	1.00	2.28 ± 0.38	1.00	(P = 1.00)	(P = 1.00)
373-K1 (Opaque orange)	UF	1258 ÷ 134	10.14	85.9 ± 2.8	1.00	80.6 ± 2.9	1.00	217 ± 20	48 ± 15
	F	219 ÷ 164	10.14	3.84 ± 0.91	1.00	3.84 ± 0.91	1.00	(P = 1.00)	(P = 1.00)
353-F2 (translucent white)	UF	268 ÷ 143	12.76	9.34 ± 0.93	1.00	7.51 ± 1.17	1.00	25.7 ± 4.1	N.D.
	F	1550 ÷ 1564	12.76	N D.	0.06	N.D.	0.01	(P = 1.00)	(P = 0.47)
353-F5 (translucent white)	UF	214 ÷ 123	12.28	8.61 ± 1.00	1.00	6.78 ± 1.23	1.00	23.1 ± 4.2	N.D.
	F	127 ÷ 128	12.28	N D.	0.30	N.D.	0.05	(P = 1.00)	(P = 0.47)

* Q – quartz; H – hornblende; F, P, K – feldspar; B – biotite; V – biotite-vermiculite; M – muscovite.
† UF – unfiltered; F – filtered.
‡ Probability of counting rate exceeding critical level for detection.
§ P – Probability of activity exceeding zero.
N.D. – Not detected.

Table 2. Continued

Sample number	Type of count	Counting rate (counts ÷ min)	Weight (mg)	Corrected counting rate (cpm/100 mg)	Probability of detection	Net counting rate (cpm/100 mg)	Probability of detection	^{137}Cs (dpm/100 mg)	^{90}Sr (dpm/100 mg)
353-F7 (translucent white)	UF	508 ÷ 145	12.90	21.9 ± 1.2	1.00	20.0 ± 1.4	1.00	68.8 ± 5.3	N.D.
	F	149 ÷ 159	12.90	N.D.	0.10	N.D.	0.01	(P = 1.00)	(P = 0.41)
373-P1 (translucent white)	UF	627 ÷ 150	12.88	27.2 ± 1.3	1.00	25.3 ± 1.5	1.00	86.9 ± 5.7	N.D.
	F	133 ÷ 140	12.88	N.D.	0.14	N.D.	0.01	(P = 1.00)	(P = 0.39)
373-F3 (translucent white)	UF	564 ÷ 166	13.13	20.7 ± 1.1	1.00	18.9 ± 1.3	1.00	64.7 ± 4.9	N.D.
	F	636 ÷ 652	13.13	N.D.	0.07	N.D.	0.01	(P = 1.00)	(P = 0.42)
010:9-F1 (translucent white)	UF	226 ÷ 158	12.08	6.19 ± 0.82	1.00	4.37 ± 1.09	1.00	14.8 ± 3.7	N.D.
	F	123 ÷ 127	12.08	N D.	0.21	N.D.	0.03	(P = 1.00)	(P = 0.48)
353-F3 (opaque white)	UF	693 ÷ 184	12.14	25.4 ± 1.2	1.00	23.6 ± 1.4	1.00	80.1 ± 5.3	N.D.
	F	857 ÷ 794	12.14	1.10 ± 0.34	0.83	N.D.	0.09	(P = 1.00)	(P = 0.40)
373-P2 (opaque white)	UF	1917 ÷ 151	18.38	65.4 ± 1.6	1.00	63.5 ± 1.8	1.00	215 ± 19	14 ± 15
	F	233 ÷ 160	18.38	2.44 ± 0.52	1.00	1.58 ± 0.90	0.61	(P = 1.00)	(P = 0.83)
353-B1	UF	2046 ÷ 127	13.40	115.1 ± 2.9	1.00	107.8 ± 2.9	1.00	354 ± 19	16 ± 12
	F	169 ÷ 136	13.40	2.21 ± 0.73	0.98	2.21 ± 0.73	0.98	(P = 1.00)	(P = 0.91)
373-B1	UF	5271 ÷ 85	15.76	389.1 ± 6.2	1.00	381.9 ± 6.2	1.00	1291 ± 42	51 ± 14
	F	331 ÷ 157	15.76	7.36 ± 0.75	1.00	7.36 ± 0.75	1.00	(P = 1.00)	(P = 1.00)
353-V1	UF	6215 ÷ 173	11.56	304.9 ± 5.1	1.00	302.3 ± 5.1	1.00	799 ± 40	208 ± 26
	F	392 ÷ 131	11.56	17.7 ± 1.3	1.00	16.0 ± 1.6	1.00	(P = 1.00)	(P = 1.00)
373-V1	UF	5228 ÷ 45	17.64	655 ± 10	1.00	652 ± 10	1.00	2266 ± 72	72 ± 23
	F	455 ÷ 138	17.64	13.33 ± 0.89	1.00	11.7 ± 1.2	1.00	(P = 1.00)	(P = 1.00)
373-M1	UF	588 ÷ 137	7.00	51.6 ± 2.7	1.00	44.8 ± 2.8	1.00	144.4 ± 9.8	N.D.
	F	179 ÷ 157	7.00	2.8 ± 1.3	0.87	N.D.	0.30	(P = 1.00)	(P = 0.31)

Pink and Orange Feldspars

The common feature of grains in the pink and orange group was the presence of small, bright red hematite inclusions which are always visible in thin section. The combined amount of pink and orange feldspar comprised less than 10% of the total segregated feldspar. About 50% of the opaque pink and orange grains were actually rock fragments which contained trace to minor amounts of mafic minerals.

Thin-section analysis of sample 353-F4 (Table 2) showed translucent pink grains to be largely microcline. One quarter of the grains were polysynthetically twinned in the manner of plagioclase (chiefly oligoclase, with some albite). Thin-section analysis also showed opaque grains to be extensively altered. Identification of the altered grains was very difficult, although at least some were sodic plagioclase.

The degree of translucency or opacity of pink and orange feldspar grains reflected the presence of sericite and very fine grain, white to orange-brown alteration products that could not be positively identified. The degree of sericitization paralleled the development of the other alteration products. In moderately altered samples, the very fine alteration products were localized along, but not restricted to, cleavage planes and microfractures. It is thought that the orange-brown product may be either minute inclusions of exsolved hematite or iron oxide alteration products of hematite.

White Feldspars

The term "white feldspar" refers to grains that ranged in translucency from being opaque to almost transparent and colourless. As in the pink and orange feldspars, the degree of translucency or opacity depended upon the amount of sericite and fine-grained, unidentified alteration products. In total, less than 10% of the feldspar grains were opaque white.

Thin-section analysis of the white feldspars indicated that oligoclase was the predominant mineral, and determination of optic axial angle and sign suggested a composition of about An_{20}. Some microcline was present in all thin sections (although it decreased in abundance in samples exhibiting a greater degree of alteration), as was albite. Alteration products obscured most optical properties of the opaque grains. The plagioclase was not zoned and some anti-perthite was observed. A few grains in every thin section contained inclusions of hematite, although none of the grains exhibited the characteristic staining of pink and orange feldspar. Epidote was identified in some grains, indicating that saussuritization had occurred.

Available evidence indicates that the alteration products in all feldspars observed in thin section were produced by metamorphic or hydrothermal processes and not by recent weathering. The evidence includes the presence of saussurite and sericite and indistinct boundaries separating altered and unaltered feldspar. Distinct boundaries are believed to be characteristic of alteration by weathering processes (Rodgers and Holland, 1979; Walker *et al.*, 1978).

SEM-EDS Analysis of Feldspars

The SEM-EDS analysis of the surfaces of the orange and pink feldspar grains showed a wide variation in elemental composition suggesting albite, perthite and microcline. Out of a total of five white feldspar grains that were similarly examined, four grains appeared to contain Ca in greater amounts than Na and K. This observation was not consistent with the results of the thin-section analysis of white feldspar segregates, and it is not known whether the apparent predominance of Ca at the mineral surface is the result of X-ray source geometries or of true chemical alteration. The feldspar matrix spectra were not useful in identifying alteration products, but all grain surfaces contained Fe.

Several feldspar grains that had been segregated from core RA3 samples and from other cored sediments in the lower Perch Lake Basin were observed by SEM-EDS, but only one grain surface was noted to be extensively etched. None of the grain surfaces of feldspars obtained from core RA3 appeared to be significantly smoothed by precipitation coats.

All of the feldspar surfaces that were examined by SEM were littered with adhering particles, and maximum coverage of about 5% of the total surface area occurred in sample 010:9-F1. Grain matrix spectra interfered with the particle spectra, and so it was difficult to determine the particle compositions. Given the wide variation in compositions that was observed, the simplest interpretation is that most particles were glacial rock flour. Many particles, however, had a composition very similar to that of the matrix to which they were adhering, suggesting a surface splintering origin.

Levels of ^{137}Cs and ^{90}Sr in Feldspars

The levels of ^{137}Cs and ^{90}Sr sorbed by pink and orange feldspar segregates ranged from below detection limits to 217 ± 20 dpm/100 mg and 48 ± 15 dpm/100 mg, respectively (Table 2). The only apparent difference between opaque orange samples from the two core intervals was the presence of a greater percentage of red-orange grains

in sample 373-K1 (30% vs. 10% in sample 353-K1). The radioisotope ^{90}Sr was possibly detected in only one white feldspar segregate (373-P2) but all eight white feldspar segregates contained ^{137}Cs (23 ± 4 to 215 ± 19 dpm/100 mg).

The association of ^{90}Sr with pink and orange feldspars may be due to primary grain mineralogy or, more probably, the presence of oxides. The grains in the translucent pink segregates were predominantly microcline with minor plagioclase, whereas the white segregates that did not contain detectable amounts of ^{90}Sr were mostly plagioclase with minor microcline. A greater abundance, or degree of alteration, of included hematite in the coloured samples suggests that much of the sorption may be due to iron oxides.

Regardless of colour, the opaque and more highly altered feldspar segregates contained greater levels of sorbed ^{137}Cs than the translucent segregates (Table 2). The significance of the very fine-grained unidentified alteration product in ^{137}Cs sorption could not be determined, but its role could be considerable if it contained sericite and zeolite. On the basis of structure, sericite would be expected to retain trace levels of ^{137}Cs as an interlayer cation. Some zeolites have a high sorptive capacity for Cs, and are strongly selective for Cs compared with Sr (Berak, 1963). Regardless of its origin, plagioclase usually contains some sericite (Moorehouse, 1959), and so it is expected that plagioclase in most sediments will sorb and retain some Cs.

The retention of Cs by feldspar grains in the lower Perch Lake Basin was first noted by Evans (1954), who reported that significant levels of ^{134}Cs and ^{137}Cs were not readily desorbed by H_2O_2, $CaCl_2$ or Na_2CO_3 solutions.

Micas

Biotite grains were segregated on the basis of their black colour and characteristic platy shape. Most grain surfaces had a dull appearance in comparison with freshly cleaved (001) planes, which had a vitreous lustre. The dull lustre is undoubtedly produced by chemical alteration of the grain surfaces. Biotite-vermiculite (X-ray diffraction analysis confirmed vermiculite) was segregated on the basis of surface alteration colour. Biotite-vermiculite surfaces ranged from dark green to bronze, and single grains often displayed the full range of colour.

Muscovite grains separated from the glass block site sediments were transparent to translucent, depending upon the presence of minute inclusions or the development of alteration products. Sediment sample RA5:353 did not contain sufficient muscovite in the 0.42- to 0.50-mm grain size fraction to permit preparation of a muscovite segregate.

With one exception, 373-V1, only a few grains in all mica segregates contained visible trace patches of iron oxide. Sample 373-V1 exhibited an iron oxide coating on about 5% of its total surface area.

Unlike almost all other mineral segregates that were prepared for counting, a difference was observed between biotite and biotite-vermiculite grains that were separated from the two RA5 sampling intervals. Grains obtained from RA5:373 were altered to a greater degree, as shown by a greater abundance of dark green biotite and by a greater intensity of bronze colouration in the biotite-vermiculite. Also, biotite and biotite-vermiculite grains in this sample were present in approximately equal abundance, whereas biotite was much more abundant than biotite-vermiculite in sample RA5:353. The greater degree of alteration shown in RA5:373 samples is considered to be the result of a primary sediment sorting mechanism. Patterson (1979) has noted that active weathering of biotite in the lower Perch Lake Basin appears to be restricted to the upper 1 to 2 m of the section. Moreover, fine grains of biotite in the two core RA5 intervals were not altered more than the coarse grains, as would be expected if active weathering had been responsible.

SEM-EDS Analysis of Micas

The X-ray spectra for the biotite and biotite-vermiculite surfaces indicated that with alteration grain surfaces become relatively depleted in Cl and K and enriched in Ca, Mg, Fe, Ti and Cr, consistent with a transformation from biotite to vermiculite. With few exceptions, the X-ray spectra of particles on biotite and biotite-vermiculite grain surfaces were very similar to the corresponding matrix spectra. Photomicrographs indicated that these particles originated from the grain surfaces by some physical process of splintering. Also, the photomicrographs showed that the grains have begun to open along cleavage planes. These openings, as well as the presence of particles derived from the mineral surface, have the potential to increase the effective surface area and sorptive capacity of biotite and biotite-vermiculite grains beyond what would be expected from considerations of grain size and shape alone.

Muscovite grains did not have visible surface coatings of Fe oxide, yet matrix spectra indicated that a minor amount of Fe was present. At a magnification of × 5000, the muscovite grain edges appear to be partially opened, but not to the extent observed in the biotite and biotite-vermiculite.

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Levels of ^{137}Cs and ^{90}Sr in Micas

Levels of ^{137}Cs sorbed by biotite-vermiculite are approximately twice the levels of ^{137}Cs sorbed by biotite in each stratigraphic interval (Table 2). For sediment sample RA5:373, the biotite segregate sorbed more ^{137}Cs (1291 dpm/100 mg) on a weight basis than the muscovite segregate (144 dpm/100 mg). This result is consistent with the observations that the alteration of biotite, including opening of grain edges, has proceeded to a greater extent than muscovite. Sawhney (1970) has shown that the sorptive properties of 2:1 phyllosilicates are determined to a significant extent by the areas of "frayed" mineral edges (edge-expanded interlayers which form so-called wedge sorption zones). The significance of alteration was further demonstrated by the results for sample 373-V1, which contained the most visibly altered grains and had the highest level of ^{137}Cs (2266 ± 72 dpm/100 mg).

Both biotite and muscovite may weather to vermiculite with the loss of interlayer K and deposition of Al hydroxide interlayers. Although it is generally thought that large mica grains initially weather from grain edges towards grain centres, Jackson et al. (1973) have noted that cation diffusion and deposition of hydrous Fe and Al oxides along cleavage planes are facilitated by numerous grain microfractures and pores. Expanded vermiculite interlayers also can readily accommodate hydrated alkaline earth cations such as Sr.

Biotite-vermiculite grains from the glass block site appear to have a greater ^{90}Sr retention capacity than biotite grains (Table 2). A significant correlation between the degree of biotite weathering or alteration to vermiculite and the amount of sorbed ^{90}Sr is evident only for the segregates obtained from sediment sample RA5:353 (16 ± 12 dpm/100 mg for biotite vs. 208 ± 26 dpm/100 mg for biotite-vermiculite). Oxides may be responsible for some of the differences between the biotite and biotite-vermiculite segregates. However, segregate 373-V1, which contained the most altered and iron-rich grains, did not contain the highest level of sorbed ^{90}Sr. These observations suggest that other mechanisms of retention may be important in the micas. Specifically, it is possible that non-uniform interlayer sorption of ^{137}Cs has caused nonuniform collapse of interlayers and isolation of regions containing alkaline earth interlayer cations such as Sr. This mechanism of isolation has been described by Douglas (1977).

Activity of Sieved Sediment Fractions

Sample RA5:353 was sieved into several phi interval size fractions that were split mechanically and counted. A stratigraphically equivalent interval from core RA3 (346–367 cm) was also sieved to produce corresponding grain size fractions for measuring blank counting rates. Net counting rates and levels of ^{137}Cs and ^{90}Sr for the sieved fractions of RA5:353 are reported in Table 3.

Table 3. Sample RA5:353 Radioactive Contamination of Whole Sediment and Sieved Fractions

Size range (mm)	Type of count*	Net counting rate (cpm/100 mg)	Probability of detection†	^{137}Cs ‡ (dpm/100 mg)	^{90}Sr ‡ (dpm/100 mg)	Sample weight (%)	Weighted net counting rate (cpm/[wt. %] × [100 mg])
Whole sediment	UF	11.83 ± 0.37	1.00	52.7 ± 3.1 (P = 1.00)	2.1 ± 1.7 (P = 0.88)	100	11.83 ± 0.37
	F	0.25 ± 0.10	0.84				
>0.5	UF	35.95 ± 0.66	1.00	137.6 ± 5.4 (P = 1.00)	23.2 ± 2.6 (P = 1.00)	0.261	0.09 ± 0.00
	F	1.67 ± 0.14	1.00				
0.25–0.5	UF	14.40 ± 0.41	1.00	—	—	26.972	3.88 ± 0.11
0.125–0.25	UF	10.56 ± 0.18	1.00	46.1 ± 3.2 (P = 1.00)	2.7 ± 2.2 (P = 0.88)	63.115	6.66 ± 0.11
	F	0.26 ± 0.12	0.84				
0.0625–0.125	UF	13.50 ± 0.36	1.00	—	—	9.383	1.27 ± 0.03
<0.0625	UF	32.33 ± 0.76	1.00	147.2 ± 7.8 (P = 1.00)	1.6 ± 4.6 (P = 0.64)	0.268	0.09 ± 0.00
	F	0.46 ± 0.26	0.70				
Total of sieved fractions						99.999	11.99 ± 0.16

* UF — unfiltered; F — filtered.
† Probability of net counting rate exceeding critical level for detection.
‡ P — probability of activity exceeding zero.

Although the silt and clay fraction and the coarse and very coarse sand fraction (grain size greater than 0.5 mm) each comprise less than 0.3% by weight of sample RA5:353, their counting rates on a weight basis were three times greater than those of the intermediate size fractions. This observation is consistent with the results for the mineralogic segregates and other studies of lower Perch Lake Basin sediments (Jackson and Inch, 1980; Patterson and Spoel, 1981) and has been attributed to a relatively higher abundance of micaceous minerals in both the coarsest and finest fractions and the greater specific surface area of the finest fraction. Microscopic examination of grain size fractions of sediments from the glass block site indicates a similar distribution of micaceous minerals. An increased abundance of biotite grains in the finer fractions probably accounts for the higher blank counting rate (4.3 cpm/ 100 mg) of the silt and clay size fraction of RA3:346.

Relatively high levels of ^{137}Cs occur in both the coarsest and the finest fractions; ^{90}Sr, however, is enriched only in the coarse segregate. The reason for relatively low levels of ^{90}Sr in the fine fraction is not clear because sieved samples of silt and clay laminae (grain size less than 0.0625 mm) from core RA5 generally contained, when detected, significantly more ^{90}Sr than ^{137}Cs. Results have been reported (Jackson and Inch, 1980; Catto, 1978) which show that some silt and clay laminae in the lower Perch Lake Basin contain more interstratified mica-vermiculite and chlorite than the silt and clay size fraction of sandy sediments. This mineralogical difference, however, does not readily explain the distribution of ^{90}Sr contamination with grain size.

DISCUSSION

Distribution of Retained ^{137}Cs

For each of the two sediment samples, RA5:353 and RA5:373, the complete series of mineral segregates can be ranked on a weight basis according to the measured amount of sorbed ^{137}Cs (Table 2) as follows:

biotite-vermiculite >
biotite >
opaque or altered feldspar >
muscovite >
hornblende ≥
translucent feldspar ≥
quartz.

This ranking of ^{137}Cs mineral sorbents is the first to be determined for all the abundant minerals in lower Perch Lake Basin sediments and is similar to a partial

ranking, reported by Evans (1954), of chemically treated (H$_2$O$_2$, CaCl$_2$ and NaCO$_3$) grains, i.e., feldspar > hornblende > quartz (possibly contaminated with feldspar).

Biotite and biotite-vermiculite are only relatively minor components in the lower Perch Lake Basin sediments but they play a disproportionate role in the sorption of ^{137}Cs. As noted by Jackson and Inch (1980), the presence of biotite and biotite-vermiculite is largely responsible for the observed variation in ^{137}Cs contamination on a weight basis for different stratigraphic units, sediment sample sub-splits and grain size fractions. This is not to say, however, that the bulk of the ^{137}Cs in a whole sediment is associated with biotite and biotite-vermiculite. Jackson and Inch (1980) have suggested that most ^{137}Cs is probably sorbed by feldspar due to the greater abundance of this mineral. This suggestion is reasonable, but it may not apply to the 0.42- to 0.50-mm grain size range. In this fraction, translucent feldspar (which is the second poorest sorbent of ^{137}Cs on a weight basis and which is less sericitic than opaque feldspar) comprises over 80% of the total feldspar.

Most of the ^{137}Cs released from the blocks has been retained by sediments at the glass block site and is not electrostatically adsorbed and readily exchangeable with other cations. Nineteen years after emplacement of the blocks, 90% of the total amount of ^{137}Cs released from the glass had travelled no farther than 0.3 m (Walton and Merritt, 1979). Moreover, other researchers (Evans, 1954; Jackson and Inch, 1980) report experimental results which indicate that significant amounts of ^{137}Cs cannot be desorbed easily from contaminated lower Perch Lake Basin sediments.

Distribution of Retained ^{90}Sr

The retention of ^{90}Sr on the sediments at the glass block site has been much less significant than the retention of ^{137}Cs. After 11 years of glass block emplacement, a ^{90}Sr-contaminated ground-water plume could no longer be clearly defined because of advection and dispersion of ^{90}Sr from the site (W.F. Merritt, unpublished data). Only 9 of 20 mineral segregates in the present study contained detectable amounts of ^{90}Sr, and the highest measured level was an order of magnitude less than the highest level of ^{137}Cs (Table 2). Nevertheless, the mineral segregates can be ranked on a weight basis according to the amount of retained ^{90}Sr as follows:

biotite-vermiculite ≥
biotite and hematite-bearing feldspar >
all other mineral segregates (quartz, white feldspar, hornblende and muscovite).

As with ^{137}Cs sorbents, this ranking for ^{90}Sr is the first to be determined for all the most abundant minerals in lower Perch Lake Basin sediments.

Mineral Grain Characteristics

Fewer mineralogic subdivisions can be defined in the ranking of ^{90}Sr sorbents than for [137]Cs. Analysis of the data suggests that the difference is due to ^{90}Sr sorption being more strongly dependent on sediment characteristics that are less mineral specific. In particular, retention of ^{90}Sr appears to be related to the presence or absence of Fe and possibly Mn and Al oxide coatings which can be present on any of the mineral grains. For example, the core RA5 segregate of quartz that contained measurable ^{90}Sr also had a visible trace amount of Fe oxide on grains. Similarly, the ^{90}Sr-bearing feldspars were those coloured by hematite or containing very fine grains of either hematite or hematite alteration products. The apparent correlation between retained ^{90}Sr and metal oxides has been supported recently by the results of laboratory research conducted with contaminated lower Perch Lake Basin sediments (Jackson and Inch, 1983). The data show that there is a strong correlation between the amount of ^{90}Sr and the amount of Fe, Mn and Al that can be extracted with acidified 0.1 M hydroxylamine hydrochloride. Iron oxide coatings, as observed with the reflecting microscope, were not amenable to characterization with the SEM-EDS technique.

Two other characteristics that could be common to more than one mineral phase and that could potentially cause retention are microfractures and glacial rock flour coatings. Grain microfractures could cause the non-equilibrium sorption of trace amounts of solutes as a result of diffusion-controlled processes. Krinsley and Hyde (1971) have noted that quartz grains from glacial environments are characterized by a great number of cracks (0.1 μm to 0.25 μm wide) which are observable in the cathodoluminescence emission mode of scanning electron microscopes. Lower Perch Lake Basin grains have undoubtedly been fractured during glacial transport. However, since some quartz and feldspar segregates showed non-detectable levels of ^{137}Cs and/or ^{90}Sr, it appears that either microfractures are not common, or if they are abundant, they do not result in significant retention.

The SED-EDS analysis indicated that most of the adhering particles on quartz, feldspar and hornblende grains are likely glacial rock flour. It is thought that the adhering particles did not significantly influence counting rates because the feldspar segregate (010:9-F1) containing the greatest number of adhering particles had the lowest counting rate of all the feldspar segregates. Also, as with micro-

fractures, the non-detectable activity levels of some segregates suggest that this factor is not significant.

[90]Sr Retention

Previous work has shown that much, but not all, sorbed ^{90}Sr can be readily desorbed from contaminated lower Perch Lake Basin sediments in laboratory experiments (Ophel et al., 1972; Ophel, 1973; Jackson and Inch, 1980; Pickens et al., 1981; Killey, 1982). In addition, discrepancies between the predicted and measured patterns of distribution of sorbed ^{90}Sr at the glass block site in 1963 indicate that some non-equilibrium sorption occurred (Walton and Merritt, 1979; Melnyk et al., 1984).

Calculations incorporating data obtained in this study suggest that the ^{90}Sr which still remains at the glass block site represents a proportion of the originally adsorbed ^{90}Sr that was not readily exchangeable with other cations. Measured levels of retained ^{90}Sr can be compared with predicted levels of sorbed ^{90}Sr calculated from values of K_d^{Sr} determined by Patterson and Spoel (1981) and from the historical concentrations of ^{90}Sr in ground water at the glass block site. The relationship among these variables may be written as

$$(XSr) = K_d^{Sr} (Sr) \qquad (1)$$

where (XSr) is the quantity of Sr sorbed per gram of mineral sorbent and (Sr) is the equilibrium concentration per millilitre of solution.

Patterson and Spoel (1981) used a laboratory batch test technique to determine the values of K_d^{Sr} for various mineral segregates prepared from uncontaminated sediment in core RA1. Segregates weighing between 63 and 98 mg were separated by hand and were batched for 48 h with water obtained from piezometer RAb-2 that had been spiked with a trace amount of [85]Sr. The measured values of K_d^{Sr} for different mineral segregates are summarized in Table 4. Table 5 lists the amounts of sorbed ^{90}Sr that would be predicted using Equation 1, the reported K_d^{Sr}

Table 4. Sample RA1:330 Measured K_d^{Sr}'s of Mineral Segregates

Mineral segregate	K_d^{Sr*} (mL/g)
Quartz with minor feldspar	0.4
Mixed feldspar (predominantly white)	4.7
Biotite	3.7
Biotite altered to vermiculite	37
Muscovite	2.6

*Patterson and Spoel (1981).

Table 5. Measured vs. Predicted Amounts of Sorbed ^{90}Sr for Mineral Segregates

Mineral segregate	Measured amount (dpm/100 mg)*	Predicted amount from ground-water concentration of 290 dpm/mL† (dpm/100 mg)	Predicted amount from ground-water concentration of 5.3 X 10^{-1} dpm/mL‡ (dpm/100 mg)
Quartz	N.D.	12	0.02
Quartz with trace Fe oxide	15 ± 10	—	—
Mixed feldspar (largely white)	—	140	0.25
Translucent pink and opaque orange feldspar	N.D.; 29.7 ± 3.6 32 ± 6: 48 ± 15	—	—
Translucent and opaque white feldspar	N.D.; 14 ± 15	—	—
Biotite	16 ± 12; 51 ± 14	110	0.20
Biotite-vermiculite	208 ± 26; 72 ± 23	1100	2.0
Muscovite	N.D.	75	0.14

* N.D. — Not detected. One measurement obtained per reported value except quartz (N.D. — 2 measurements) and translucent and opaque white feldspar (N.D. — 6 measurements).
† Average ground-water concentration in 1960.
‡ Average ground-water concentration in 1977.

values (Table 4) and ^{90}Sr ground-water concentrations of 290 dpm/mL (1.3 x 10^{-1} μCi/L) and 5.3 x 10^{-1} dpm/mL (2.4 x 10^{-4} μCi/L). The high concentration value is the average concentration of ^{90}Sr recorded for ground water in 1960 in a piezometer located 1.8 m downgradient from the blocks (Merritt, 1976). The low value is the average concentration of ^{90}Sr in water obtained during 1977 (Walton and Merritt, 1979) and is representative of average annual ^{90}Sr concentrations since 1966. Table 5 also lists the measured amounts of sorbed ^{90}Sr that were determined during this study (Table 2).

Strict comparisons, of course, cannot be made between the measured amounts of ^{90}Sr sorbed by core RA5 segregates and the amounts calculated using Equation 1 and K_d^{Sr} and ^{90}Sr concentration data. The sediments from core RA5 (located 0.4 m downgradient from the glass blocks) probably were exposed to higher levels of ^{90}Sr than were measured in the sampler (located 1.8 m downgradient) in 1960. Also, the range in grain size of batch test segregates was not reported by Patterson and Spoel (1981). A visual inspection of the batch test grains at the time of their preparation indicated that they were, on average, smaller than the grains segregated from core RA5, and so the batch test grains would be expected to sorb a greater amount of ^{90}Sr on a unit weight basis.

The measured amounts of ^{90}Sr on the mineral segregates are much lower than the quantities predicted using the K_d^{Sr} results and the 1960 ground-water concentration value of 290 dpm/mL ^{90}Sr (compare columns 2 and 3 in Table 5).

Since the concentration value for ground water is probably a conservative estimate of the maximum level of exposure experienced by the sediments, the data suggest that for biotite and biotite-vermiculite less than 20% of the ^{90}Sr which was likely sorbed soon after emplacement and leaching of the blocks is still retained.

In comparison with the results based on the average ^{90}Sr concentration in 1960, measured quantities of retained ^{90}Sr are much higher than those calculated using the 1977 ground-water ^{90}Sr concentration of 5.3 x 10^{-1} dpm/mL (compare columns 2 and 4, Table 5). These large differences, particularly considering that the sediments had been exposed to the dilute ground waters for over 10 years, emphasize the conclusion that the remaining ^{90}Sr on the quartz (with trace Fe oxide), pink and orange feldspar, biotite and biotite-vermiculite is very strongly sorbed and not readily exchangeable.

The results of the study are consistent with observations by other researchers that a variety of sites, in terms of degree of reversibility, exist on C.R.N.L. sediments for sorption of ^{90}Sr. Whereas all ^{90}Sr sorbed during three-day batch experiments is readily exchangeable (Killey, 1982), only 20% to 50% of ^{90}Sr sorbed during contact times of approximately 20 to 25 years can be removed with CaCl$_2$ or SrCl$_2$ solution desorbing treatments (Jackson and Inch, 1983 and 1980; Killey, 1982). Killey (personal communication) also has noted that non-equilibrium, slow desorption of ^{90}Sr results in higher apparent K_d^{Sr} values in older portions of a contaminant plume. The work of these researchers

indicates that most of the ^{90}Sr retained by the sediments is associated with Fe, Mn, and Al oxides. Moreover, the results of modelling the glass block site soil survey data obtained in 1963 are improved when two sorption mechanisms are incorporated (Melnyk et al., 1984).

CONCLUSIONS

The following conclusions have been reached:

1. The radionuclide ^{137}Cs has been strongly retained by the sediments and is not readily exchangeable with other cations. The major mineral phases that retain the cesium are the layer silicates, principally biotite-vermiculite and biotite. Significant retention also occurs on feldspars which contain alteration products such as sericite.

2. Compared with ^{137}Cs, ^{90}Sr is much less strongly retained and its distribution in the sediment is less mineral-specific. The most effective mineral-sorbent phases in the sediments are biotite-vermiculite and biotite. Results of this and other studies indicate that a spectrum of ^{90}Sr sorption sites, ranging from readily reversible to very slowly reversible, exists on the sediment. The retained ^{90}Sr is associated in significant proportion with oxides, particularly of iron.

ACKNOWLEDGMENTS

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