

INTERNATIONAL GROUNDWATER SYMPOSIUM
ON
HYDROGEOLOGY OF COLD AND TEMPERATE CLIMATES
AND
HYDROGEOLOGY OF MINERALIZED ZONES

P R O C E E D I N G S

Editor: C.L. Lin

Halifax, Nova Scotia, Canada
May 1-5, 1988

Sponsored by
ATLANTIC CANADA REGION

The International Association of Hydrogeologists
Canadian National Chapter



VARIATIONS IN NATURAL-DECAY SERIES DISEQUILIBRIUM
ALONG GROUNDWATER FLOWPATHS

Kevin A. Morin

Morwijk Enterprises
Suite 1706L, Laurier House,
1600 Beach Avenue,
Vancouver, British Columbia B6G 1Y6
Canada

and

John A. Cherry

Institute for Groundwater Research
University of Waterloo,
Waterloo, Ontario N2L 3G1 Canada

Paper presented at the International Groundwater Symposium of
the International Association of Hydrogeologists, Canadian National
Chapter, Atlantic Region, Halifax, Nova Scotia, May 1-5, 1988

Variations In Natural-Decay-Series Disequilibrium Along Groundwater Flowpaths

Kevin A. Morin
Morwijk Enterprises
Suite 1706L, Laurier House,
1600 Beach Avenue,
Vancouver, British Columbia V6G 1Y6
Canada

John A. Cherry
Institute for Groundwater Research
University of Waterloo,
Waterloo, Ontario N2L 3G1
Canada

ABSTRACT

Activity ratios of radionuclides in groundwater are often not equal to 1.00, indicating isotopic disequilibrium is common, and variations in the ratios along groundwater flowpaths indicate the operation of solid-liquid geochemical and decay mechanisms, such as sorption, co-precipitation, and alpha recoil. A review of non-mining-related studies shows that activity ratios in the ^{238}U , ^{232}Th , and ^{235}U decay series can differ greatly among sites and may or may not show trends along flowpaths. The reviewed uranium-mining-related studies, predominately in and near tailings and waste-rock dumps, also reveal a wide range of values for activity ratios among sites, but trends along the flowpaths of 100-1000 meters in length are often apparent. A field study presented here examines in detail the variations in activity ratios in the ^{238}U , ^{235}U , and ^{232}Th series along a flowpath of 21 meters, monitored by seven piezometers in the vicinity of a uranium-tailings impoundment. The $^{234}\text{U}/^{238}\text{U}$ ratio, for example, increases from 1.23 to 3.92 and $^{227}\text{Th}/^{227}\text{Ac}$ decreases from 1.88 to 1.26 along the flowpath. Also, the isotopes of a particular element display different aqueous behaviors. For example, ^{230}Th steadily decreases along the flowpath from 1370 to <19 mBq/L, whereas ^{228}Th initially increases from 130 to 190 mBq/L then decreases to <19 mBq/L. The variations in radionuclide activities and the resulting activity ratios along groundwater flowpaths are controlled by the particular suite of geochemical and decay mechanisms that regulates each isotope of each element.

1. INTRODUCTION

In the vicinity of uranium-ore deposits, the aqueous migration of radioactive elements of contrasting mobilities is commonly observed. This migration in water produces geochemical halos in surrounding soil and rock that can be detected by soil-rock analyses. The differing mobilities in water of radionuclides and the differing degree of solid-liquid interactions among radionuclides commonly result in decay-series disequilibrium in both the water and rock. A man-induced analog of this process can be found in the contaminant hydrogeology of uranium-mill tailings.

Uranium-mill tailings can represent an extreme example of a radioactive deposit in which (1) the ore is fine-grained and unconsolidated, (2) the porewater contains strong leach chemicals, and (3), because most of the uranium is removed during milling, decay-series disequilibrium initially exists in the tailings solids and porewater. The contaminated porewater, which is commonly replenished by infiltration of precipitation and/or mill effluent, often drains downward into underlying groundwater flow systems (Morin 1988 A and B, these proceedings; Morin and Cherry, submitted). Consequently, radionuclide-laden groundwater seepage from tailings impoundments provides an opportunity to examine radionuclide migration and deviation from decay-series equilibrium.

This paper presents measurements of aqueous disequilibrium in the three natural decay series at uranium-mining and non-uranium-mining sites. A detailed field study of tailings seepage presented here particularly shows the notable change in the degree of disequilibrium along a flowpath of approximately 21 meters.

2. PREVIOUS STUDIES OF AQUEOUS DISEQUILIBRIUM ALONG A FLOWPATH

Although there is an abundance of published data on disequilibrium in soil, rock, and water, there are fewer studies which trace disequilibrium along a groundwater flowpath. This section reviews published studies of disequilibrium along flowpaths and in some cases includes a re-interpretation of data by defining flowpaths at a few sites.

The extant natural decay series are named for the primary long-lived parent radionuclide: ^{238}U , ^{232}Th , and ^{235}U . The ^{238}U series, which is the series most often examined in natural environments, begins with ^{238}U and ends with stable ^{206}Pb . Intermediate radionuclides which are commonly examined include ^{234}U , ^{230}Th , ^{226}Ra , ^{222}Rn , and ^{210}Pb . The variations in ratios, such as in $^{234}\text{U}/^{238}\text{U}$ and $^{226}\text{Ra}/^{230}\text{Th}$, along a groundwater flowpath reflect the relative effects of radioactive decay (often over relatively large times and distances) and of water-rock interaction (over short and long times and distances) on the two radionuclides in the ratio. An understanding of the decay series and of the complex behavior of radionuclides in groundwater flow systems (e.g. Morin et al., accepted, B; Davidson and Dickson, 1986) is required in order to understand the abbreviated discussions in this paper.

2.1 Non-Mining-Related Studies

In a study of the Floridan Aquifer in northern Florida, Kaufman et al., (1969) found $^{234}\text{U}/^{238}\text{U}$ activity ratios ranging from 0.50 to 1.2 at uranium concentrations of 0.21 to 3.42 $\mu\text{g}/\text{L}$ with one large value of 25.91 $\mu\text{g}/\text{L}$. Ratio values less than 1.0, indicating relative depletion of ^{234}U , were located in areas of high permeability, active groundwater circulation, and apparently greater oxidation potential allowing greater mobilization of uranium. There was an inverse relationship of the activity ratio to aqueous uranium concentration which is commonly found at other sites. No clear spatial trend in the ratio is discernible in the reported data as the groundwater moves tens of kilometers towards the ocean, possibly because of irregular karst conditions in the aquifer and the importance of local flow systems.

Although Titayeva et al. (1973) did not examine disequilibrium along a groundwater flowpath, thirteen springs and nearby locations were sampled along a 400 meter length of essentially soil-less valley wall in the Polar Ural. This study is of particular value because it examines the ^{232}Th as well as ^{238}U decay series. For Springs 1-13 which issue low-TDS, sodium-bicarbonate water, the researchers report six $^{234}\text{U}/^{238}\text{U}$ values of 0.9-1.27 and two $^{230}\text{Th}/^{238}\text{U}$ values of 0.63 and 1.11. Mean uranium and thorium concentrations were 0.08 and 0.11 $\mu\text{g}/\text{L}$, respectively. For the ^{232}Th series in Springs 1-13, four values of 7.5-17.7 are reported for $^{228}\text{Th}/^{232}\text{Th}$. Additionally, the ratio of $^{232}\text{Th}/^{230}\text{Th}$, which involves both series, varies from 0.34-1.9 in four springs. Titayeva et al. found that an increase in ratios in water (values greater than 1) generally corresponded with diminished ratios in rock.

In a study of $^{234}\text{U}/^{238}\text{U}$ disequilibrium in the Trinity Aquifer of central Texas, Kronfeld and Adams (1974) and Kronfeld (1974) reported many ratio values in groundwater ranging from 0.92 to 12.25. Corresponding uranium concentrations were around 0.01-0.13 $\mu\text{g}/\text{L}$ with two extreme concentrations of 1.870 and 15.46 $\mu\text{g}/\text{L}$ near the aquifer-recharge outcrop. Kronfeld attributed the high ratios to alpha-recoil ejection from the solid-phase uranium into groundwater during the decay of ^{238}U to ^{234}Th and subsequent rapid decay of the aqueous ^{234}Th (half-life of 24.1 days) to ^{234}Pa (h.l. of 1.2 minutes) to ^{234}U in the groundwater. The data can also be evaluated in terms of a groundwater flowpath. Surface waters in the recharge area were found to have relatively high uranium concentrations, probably accounting for the two extreme concentrations noted above, and activity ratios around 1-2. After tens of kilometers of subsurface flow, uranium concentrations were relatively low and activity ratios rose to 5-12 and there appears to be a trend of increasing activity ratio with further distance. Kronfeld examined no other ratios or decay series.

Osmond and Cowart (1974) described a scenario in the Carrizo Sandstone of southern Texas similar to Kronfeld's (1974) studies. Groundwaters near the recharge outcrop had $^{234}\text{U}/^{238}\text{U}$ activity ratios less than 1.0 and uranium concentrations of approximately 1 ppb, and ratios rose to about 9.0 and concentrations decreased to about 0.01 ppb after flowing downdip approximately 15 kilometers. The ratio trend was attributed to alpha recoil and the concentration trend attributed to the transition from relatively oxidizing to reducing conditions. Further downdip, the activity ratio decreased to about 2.0, which Osmond and Cowart attributed to

geochemical effects rather than decay effects. Frohlich et al. (1981) simulated the differential behavior of the two uranium isotopes at this site through sorption processes.

In a sandstone aquifer in the Red Desert region of Wyoming, Osmond and Cowart (1982, p.233) indicate $^{234}\text{U}/^{238}\text{U}$ ratios lie in the range of 0.8-2.5 and uranium concentrations are around 20-300 ppb updip of a geochemical uranium barrier. Downdip of the barrier, ratios are 1.6-9.7 and uranium concentrations are less than 3 ppb.

Cowart (1980) studied variations in $^{234}\text{U}/^{238}\text{U}$ in the Edwards Carbonate Aquifer in Texas. From the recharge area and along tens of kilometers of flowpath, the activity ratio remained in the range 1.07 to 1.22 with uranium concentrations of 0.662-0.820 $\mu\text{g}/\text{L}$. Parallel to the flowpath, there is a portion of the aquifer that contains slower-moving, more-reducing, higher TDS groundwater, which had ratios of 1.62-2.66 and uranium concentrations of 0.046-0.101 $\mu\text{g}/\text{L}$. The mixing zone of the two waters had anomalous uranium concentrations of 0.071-4.59 $\mu\text{g}/\text{L}$ and the ratios ranged from 0.79 to 1.40, which Cowart attributed to changes in the location of the redox boundary rather than simple mixing of the oxidized and reduced groundwaters.

In another Texas study, in the Palo Duro Basin, Laul et al. (1985) reported analyses for radionuclides in the ^{238}U and ^{232}Th series in two brines. Approximate values for ^{238}U -series ratios were: $^{234}\text{Th}/^{238}\text{U} = 25-50$, $^{234}\text{U}/^{234}\text{Th} = 0.025-0.050$, $^{234}\text{U}/^{238}\text{U} = 1.1-1.3$, $^{230}\text{Th}/^{234}\text{U} = 0.19-1.11$, $^{226}\text{Ra}/^{230}\text{Th} = 2.0 \times 10^4 - 3.0 \times 10^4$, and $^{222}\text{Rn}/^{226}\text{Ra} = 0.8-2.4$. For the ^{232}Th series, approximate values for ratios in the two brines were: $^{228}\text{Ra}/^{232}\text{Th} = 800-2040$, $^{228}\text{Th}/^{228}\text{Ra} = 0.009-0.010$, $^{228}\text{Th}/^{232}\text{Th} = 7-23$, $^{224}\text{Ra}/^{228}\text{Th} = 90-100$, and $^{224}\text{Ra}/^{228}\text{Ra} = 0.9-1.0$.

Cowart (1981) examined the $^{234}\text{U}/^{238}\text{U}$ activity ratio and ^{226}Ra activities in groundwater in the Cambrian-Ordovician aquifers in the Tri-State region (Missouri, Kansas, and Oklahoma) of the U.S.A. In the aquifers, calcium-bicarbonate water with activity ratios generally around 7-10 and uranium concentrations generally around 0.3-0.5 $\mu\text{g}/\text{L}$ flows westward and eventually mixes with sodium-chloride, H_2S -bearing (more reducing) groundwater. Within the mixing zone, ^{226}Ra increases from around 0.05 Bq/L up to 0.32 Bq/L, uranium precipitates to levels around 0.02-0.04 $\mu\text{g}/\text{L}$, and, notably, uranium activity ratios remain essentially constant. Although the decrease in uranium concentrations is consistent with reduction in Eh, Cowart could only speculate on explanations for a steady uranium ratio and an increase in ^{226}Ra . If the uranium concentrations reported as pCi/L in Cowart's Table II essentially represent ^{238}U , then the calculated values for the $^{226}\text{Ra}/^{234}\text{U}$ ratio increase from about 0.5 to about 1.5 as the calcium-bicarbonate water flows westward and there is a sharp increase to 6-236 in the mixing zone. Under the assumption made for the calculation, this trend indicates that there is relatively strong aqueous enrichment of ^{226}Ra over ^{234}U during mixing, probably synonymous with the parallel decrease in uranium concentration.

CBCL Limited (1985) carried out a detailed study of a small, post-glacial, uraniumiferous peat deposit at the base of a granitic talus slope near Portland Creek, Newfoundland. Four piezometer nests were installed in

a line perpendicular to the talus slope and, consequently, these piezometers may lie on or near the same flowpath over a distance of 135 meters. The groundwater in the peat is low-TDS, calcium-sodium-bicarbonate water. Along the line of piezometers from the piezometer closest to the slope (P1) to the most distant piezometer (P4), pH remains relatively constant near 6 and uranium decreases by a factor of only 1.7 from 415 ug/L. For the ^{238}U series, the ratio values (many calculated here from data in their Table B-5) from P1 to P4 were found to be: 1.20 to 1.04 for $^{234}\text{U}/^{238}\text{U}$, 2.73×10^{-3} to 3.52×10^{-2} for $^{230}\text{Th}/^{234}\text{U}$, >1.4 to 0.38 for $^{226}\text{Ra}/^{230}\text{Th}$, 10.15 to 26.74 for $^{210}\text{Pb}/^{226}\text{Ra}$, and 0.50 to 0.64 for $^{210}\text{Po}/^{210}\text{Pb}$. The significant variations in $^{230}\text{Th}/^{234}\text{U}$ and $^{226}\text{Ra}/^{230}\text{Th}$ may both be attributed to an increase in the activity of ^{230}Th . The relatively high values for $^{210}\text{Pb}/^{226}\text{Ra}$ may suggest an intermediate radionuclide, perhaps ^{222}Rn gas, may have accumulated in the past. The near equilibrium of $^{210}\text{Po}/^{210}\text{Pb}$ in groundwater at the site suggests that the accumulation would have developed at least one hundred years ago, because the longest half-life which controls to time-to-equilibrium in this ratio is 25 years for ^{210}Pb . For the ^{235}U series, ^{235}U was reported as varying from 0.23 at P1 to 0.13 Bq/L at P4 and $^{231}\text{Pa}/^{235}\text{U}$ is calculated to be <0.04 .

The activity ratios at the above sites are summarized in Table 1. An obvious observation is that a large suite of activity ratios spanning a natural decay series are not commonly examined along a groundwater flowpath. Nevertheless, the available data indicate a high degree of variability exists between sites and significant changes in a particular ratio can occur along a flowpath at a site. This variability reflects the differing scales of examination among the sites and the numerous processes affecting the migration of radionuclides in groundwater (e.g. Morin et al., accepted, B). Consequently, the degree of isotopic disequilibrium in all three natural decay series during groundwater flow can be major, indicating equilibrium in a large portion of a decay series should not be expected in active flow systems.

2.2 Uranium-Mining-Related Studies

Because of the potential near-surface contamination from uranium-tailings impoundments, several detailed studies on smaller scales than the previous studies have been undertaken near impoundments. Many studies report ^{226}Ra activities because it is usually considered the most toxic component in tailings seepage, but uranium and thorium are often reported as total (sum of all isotopes of an element) concentrations on a molal basis and most other radioactive elements are rarely measured. Thus, there are several studies of major-ion chemistry and total concentrations in the vicinity of uranium tailings, but fewer provide a number of ratios along a flowpath. For uranium at least, ^{238}U may often represent total uranium (addressed in Section 3), thereby allowing the calculation of $^{226}\text{Ra}/^{238}\text{U}$ and, when ^{230}Th is available, $^{230}\text{Th}/^{238}\text{U}$.

In this section, most activity ratios have been calculated from data in the original references and, in some cases, groundwater flowpaths were defined here from information in the reference. Morin (1983), Morin (1988 A), and Morin and Cherry (submitted) summarize and further evaluate the geochemistry of many of the sites discussed below.

Before examining trends along flowpaths, the source of the seepage, the tailings impoundment itself, will be discussed. Cherry et al. (1982) reviewed several uranium-tailings sites and listed chemical analyses for tailings porewater or pond water at three sites in Wyoming, U.S.A., and one site in Ontario, Canada. At all four sites, tailings-water pH was limited to 1.8-2.7, TDS ranged 12200-18000 mg/L, and major-ion composition can generally be described as predominately Fe-H-SO_4 . If the reported " U-Nat (mg/L)" is assumed to essentially represent ^{238}U , then calculated $^{230}\text{Th}/^{238}\text{U}$ ratios are 12.29 and 23.46 for two Wyoming sites and 0.01 for the Ontario site (the Nordic Main impoundment discussed in Section 3). These Wyoming values generally agree with the ratios calculated from reportedly "typical concentrations in tailings solutions" in Table 1a of Taylor (1980): 1.00 for $^{234}\text{U}/^{238}\text{U}$ and 27.78 for $^{230}\text{Th}/^{234}\text{U}$ (=27.78 for $^{230}\text{Th}/^{238}\text{U}$). Based on available activities, two $^{226}\text{Ra}/^{230}\text{Th}$ ratios for the Wyoming sites are 0.011 and 0.058, which are over an order of magnitude higher than Taylor's typical ratio of 0.0027, and the Nordic Main site in Ontario provides a ratio of 7.37. The differences in ratios in this upper portion of the ^{238}U series between the Wyoming sites and the Nordic Main site can be mostly attributed to 4 orders-of-magnitude higher ^{230}Th activities at the Wyoming sites. The three available $^{210}\text{Pb}/^{226}\text{Ra}$ ratios in Wyoming are 0.17, 0.57, and 14.57, Taylor's typical ratio is 1.00, and the Nordic Main ratio is 12.14. For the remainder of the ^{238}U series, Taylor lists typical ratios of 1.00 for $^{210}\text{Bi}/^{210}\text{Pb}$ and $^{210}\text{Po}/^{210}\text{Bi}$, indicating rapidly attained equilibrium is often expected because of short half-lives in this portion of the series.

Because of the variability in ore characteristics passing through a mill, the variability in mill processing, and the physical and chemical variability produced during and after delivery of a slurry to an impoundment (Morin, 1988 A and B, these proceedings), the activity ratios can be expected to vary with lateral and vertical distance in an impoundment. The only study that could be located on this variability of activity ratios within tailings was that of Moffett and Tellier (1978), who examined the Nordic West Arm tailings adjacent to the Nordic Main impoundment. One "high-acid" and two "low-acid" lateral flowpaths were identified within the tailings pile. The high-acid flowpath displayed no definite trends in activity ratios along its length, but the $^{230}\text{Th}/^{238}\text{U}$ ratio was around 4-22 if total uranium represents ^{238}U , the $^{226}\text{Ra}/^{230}\text{Th}$ ratio was often around 5.8×10^{-4} - 1.7×10^{-3} , and the $^{210}\text{Pb}/^{226}\text{Ra}$ ratio varied strongly from 2.05 to 240.88. For the ^{232}Th series, the $^{228}\text{Th}/^{232}\text{Th}$ ratio was around 0.36-2.00 and ^{227}Th interference was noted for some analyses. For comparison of magnitude of two series, $^{228}\text{Th}/^{230}\text{Th}$ was in the range 0.031-0.27. Additionally, no clear trends were noted in the data for TDS or uranium along the high-acid flowpath, and it is possible that local flow systems or vertical flow may be affecting lateral trends. Along the two low-acid flowpaths, there appear to be trends of decreasing TDS, decreasing or variable acidity, decreasing or variable uranium, and relatively steady values around 0.25-0.60 for $^{210}\text{Pb}/^{226}\text{Ra}$.

Highland et al. (1981) examined the chemical composition of "tailings liquor" from one of the Wyoming ponds mentioned previously and the "average groundwater" from a point approximately 1 km downgradient from the pond. From their data, pH increases from 1.9 in the pond to 6.2 in the distant groundwater, TDS decreases from 15000 to 1820 mg/L, $^{230}\text{Th}/^{238}\text{U}$ decreases

from 12.29 to 0.076, $^{226}\text{Ra}/^{230}\text{Th}$ increases from 0.012 to 1.28, and $^{210}\text{Pb}/^{226}\text{Ra}$ decreases from 14.27 to 0.79. The change in the ratios is caused by the significant, non-proportional decrease in the activities of radionuclides, particularly a nearly 3 order-of-magnitude decrease in ^{230}Th . Hoffman and Playton (1981) examined another Wyoming tailings site and presented chemical analyses from two piezometers (5-SC and 8-SC) which are approximately 150 meters from the pond. Chemical data from these wells suggest they can effectively represent an intermediate point in the above "flowpath" of Highland et al. (1981): pH = 3.6-3.7, TDS = 15984-17970 mg/L, $^{226}\text{Ra}/^{230}\text{Th} = 0.001-0.098$, $^{210}\text{Pb}/^{226}\text{Ra} = 8.41-8.73$, and $^{210}\text{Po}/^{210}\text{Pb} = 0.0-0.33$.

Taylor and Antommaria (1978), Taylor (1980), and Haji-Djafari et al. (1979) describe a Wyoming tailings area which is sufficiently instrumented to describe a four-point flowpath:

PIEZOMETER	DISTANCE FROM TAILINGS (m)	pH	TDS (mg/L)
tailings pond	0	1.95	11810
WN-6S	0	3.00	4274
WN-14H	390	5.50	4042
WN-1HDA	1070	7.25	1395

The corresponding activity ratios are listed in Table 2 and trends are apparent for decreasing $^{230}\text{Th}/^{238}\text{U}$, generally constant $^{226}\text{Ra}/^{230}\text{Th}$, and decreasing $^{210}\text{Po}/^{226}\text{Ra}$. The sharpest change in ratios occurs between WN-6S and WN-14H where pH increases from 3.00 to 5.50.

Veska (1983) studied groundwater seepage emanating from a uranium-mine waste-rock pile near Bancroft, Ontario. The sand-aquifer flow system has three-dimensional complexities which preclude the reliable definition of a flowpath through the area, but a set of piezometers were chosen from Veska's Appendix 5.12 to depict general trends. The chosen piezometers are:

PIEZOMETER	DISTANCE FROM SOURCE (m)	pH	ELEC. COND (uS/cm)
L	0	3.7	900
GR4	9	4.6	400
GR6	12	5.0	304
M3-4.76	20	6.0	442
M4-8.83	30	6.2	305
M5-11.00	39	6.5	320
M6-14.00	47	6.8	320
M7-15.17	56	7.5	290
M8-14.59	76	7.6	300
M9-15.25	94	7.6	320

Variations in activity ratios show trends of increasing $^{234}\text{U}/^{238}\text{U}$, decreasing $^{226}\text{Ra}/^{234}\text{U}$, and initially decreasing then increasing $^{210}\text{Pb}/^{226}\text{Ra}$ (Table 2). Apparently, there are one or more mechanisms producing a notable effect in the upper portion of the ^{238}U series between GR6 and M3. The sharp change in $^{226}\text{Ra}/^{234}\text{U}$ can be attributed to the factor-of-five decrease in ^{226}Ra . The sharp rise in $^{234}\text{U}/^{238}\text{U}$ between GR6 and M3 and the continued trend of relative ^{234}U enrichment were attributed to a scenario requiring preferential leaching of ^{234}U from the waste rock in the 6+ oxidation state (UO_2^{2+}), stronger adsorption of ^{238}U (as U^{4+}) over ^{234}U (as

UO_2^{2+}) in the sand aquifer, and little redox exchange between the 4+ and 6+ states (Veska, 1983).

Table 2 summarizes the activity ratios reviewed in this section. Based on the limited data for $^{234}\text{U}/^{238}\text{U}$, there is near equilibrium (0.86-2.07) in source (tailings) water as also generally noted in Table 1. The $^{234}\text{U}/^{238}\text{U}$ ratios of Veska (1983) show a trend of increasing value along the flowpath. Below ^{234}U in the ^{238}U series, notable disequilibrium occurs, but general trends can be seen. Ratios for $^{230}\text{Th}/^{238}\text{U}$ are typically greater than 4 in tailings, but decrease to less than 0.1 along flowpaths. Values for $^{226}\text{Ra}/^{230}\text{Th}$ are variable and usually less than 0.13. The data of Veska (1983) for the ratio of $^{226}\text{Ra}/^{234}\text{U}$ which spans a wide portion of the upper ^{238}U series indicate this ratio decreases along the flowpath. The $^{210}\text{Pb}/^{226}\text{Ra}$ and $^{210}\text{Po}/^{226}\text{Ra}$ ratios are variable, but appear to be somewhat less than 1 in much groundwater seepage. There is insufficient data to evaluate the lowest portion of the ^{238}U series. Furthermore, no data could be located on variations in ratios along flowpaths for the ^{232}Th and ^{235}U series.

3. FIELD STUDY OF DISEQUILIBRIUM NEAR THE NORDIC MAIN IMPOUNDMENT, ONTARIO

In a study of groundwater seepage from the Nordic Main Impoundment (Figures 1 and 2) near Elliot Lake, Ontario, Morin (1983) identified four seepage areas around the impoundment perimeter. The major seepage area, "Seepage Area A", was studied from 1979 to 1984 and the results of yearly field monitoring and computer simulations are presented in Morin et al. (accepted, A and B) and in Morin and Cherry (submitted). In particular, Morin et al. (accepted, B) examine the effect of the various processes of sorption, mineral precipitation-dissolution, co-precipitation, aqueous speciation, and decay on radionuclide migration in Area A. Additionally, it is shown in this reference that the activity of an isotope of an element can vary over orders of magnitude if the isotope comprises an insignificant portion of the total concentration. For example, if ^{227}Th comprises 0.001% of total thorium on a molal basis, then ^{227}Th activity and concentration can increase by orders of magnitude without detectably increasing total thorium and consequently without initiating further operation of the geochemical reaction that regulates total thorium concentration.

In Seepage Area A, all of the piezometers in several multi-level piezometer nests (with the prefix "M" in Figure 3) were annually pumped for pH and specific conductance measurements and a few piezometers in each nest were selected for full chemical analysis. The contaminant plume can be traced along line A-A' in Figure 3, which therefore represents the surface trace of a groundwater flowpath. The plume is located just beneath the water table at a general depth from surface of 1-2 meters. For simplicity of discussion, one piezometer in each nest was selected as representing the center of the plume and the flowpath that passes through each of these piezometers is called the "centerline" of the plume. All groundwater samples were pumped from the piezometer through dedicated tubing, through 0.45-micrometer filters, into polyethylene bottles, and acidified to a pH of less than 1.5 with concentrated HCL. Further details on the details on the piezometers, sampling techniques, and rationale for the techniques can

be found in Morin and Cherry (in press).

The measurement of relatively low radionuclide activities in such high TDS, geochemically reactive water is difficult and a high degree of analytical error (relatively large standard deviation) can be expected (Morin and Cherry, in press). As an extreme example, ^{226}Ra in samples from one piezometer was analyzed by three laboratories and reported activities were 5150, 7890, and 780 mBq/L (part of the problem was traced to the standards). However, the natural variability along the flowpath usually exceeds the analytical variability.

The results of the 1980, 1981, and 1982 analyses for radioactive isotopes in the three natural decay series along the plume centerline are presented in Table 3. The data show significantly decreasing activities of many isotopes along the centerline for a distance of 20.8 meters, which represents a groundwater travel time of approximately 24 days at measured groundwater velocities in the area. The activities of ^{238}U are confirmed by the agreement of measured total uranium with calculated total uranium from ^{238}U in Table 3. Because of the rapid flushing rate through Area A of 24 days, decay effects are relatively negligible for many of the measured isotopes and, thus, relatively fast geochemical processes must be operating to lower the activities. Also, variations of measured activities of ^{210}Pb , ^{226}Ra , thorium isotopes, and total uranium from year to year occur, probably reflecting evolutionary changes in the tailings impoundment, the source of the plume. The decrease in activities along the centerline correspond to an increase in pH and to decreases in concentrations of the major ions, i.e. ferrous iron and sulfate, and of several other ions. By examining all potential geochemical controls on the radionuclides in Table 3, Morin et al. (accepted, B) determined that total concentrations of radium and actinium were probably regulated by co-precipitation with other minerals and that total concentrations of thorium and uranium were probably regulated by mineral precipitation-dissolution. Isotopes of an element with molal (molar in Table 3) concentrations at minor levels relative to the element's total concentration can be seen in Table 3 to deviate somewhat from the trend of total concentration. For example, ^{226}Ra , which represents total radium, decreases by a factor of 2.5 from M1 to M16 in 1982 while ^{223}Ra decreases by a factor of 1.9.

More significant deviations can be seen in ^{232}Th (major isotope) vs. ^{230}Th (minor isotope). In fact, ^{232}Th and ^{228}Th also deviate from the general trend of decreasing activities along the centerline: activities initially increase and then generally decrease. This trend is confirmed by the parallel increase then decrease in total thorium which essentially represents ^{232}Th in Area A (Table 3). The differing behavior of ^{230}Th is the result of its presence in the ^{238}U series.

Also of notable importance in Table 3 is the general dominance on a Bq/L basis of the often-ignored ^{235}U series. Although ^{235}U itself was not measured, it may occur at levels close to those of ^{227}Ac , ^{227}Th , and ^{223}Ra . If 5% of total uranium is assumed to be ^{235}U (Table 3), the resulting activities are at the general levels of the other radionuclides in the series, thereby indicating ^{235}U may represent a few percent of total uranium on a molal basis (molar basis in Table 3).

To define better the behavior of each natural decay series during groundwater flow in Area A, activity ratios are presented in Table 4 from the activities in Table 3. The activities and ratios of ^{230}Th and ^{226}Ra from M19 in 1980 are anomalous and it is not known if they are reliable or erroneous. When these intermediate radionuclides at M19 are removed from consideration by calculating $^{210}\text{Pb}/^{238}\text{U}$, a value consistent with those of M16 and M17 is obtained.

In the ^{238}U series in Table 4, the ratio of $^{234}\text{U}/^{238}\text{U}$ increases from slightly greater than 1 to nearly 4 over a flow distance of 20.8 meters, indicating the relative enrichment of ^{234}U although the activities of both isotopes have decreased. For ^{230}Th , its ratios with both uranium isotopes show an initial decrease then an increase, while its ratio with ^{226}Ra shows an initial increase then a decrease to M17. This notable disequilibrium with parent and daughter radionuclides indicates thorium is influenced in a different manner and to a different degree by its regulating geochemical mechanism than uranium and radium. When ^{230}Th is removed from consideration by calculating the $^{226}\text{Ra}/^{234}\text{U}$ ratio, values closer to equilibrium are obtained (Table 4). This suggests ^{226}Ra and ^{234}U display equivalent aqueous behavior despite a deviant ^{230}Th behavior. When ^{234}U is removed from consideration through the calculation of $^{226}\text{Ra}/^{238}\text{U}$, the resulting ratio values increase from 0.96 at M1 to 3.45 at M17. Because these values generally parallel those of $^{234}\text{U}/^{238}\text{U}$, equivalent aqueous behaviors for ^{226}Ra and ^{234}U are again indicated. The relatively small variations and lack of clear trends in $^{210}\text{Pb}/^{226}\text{Ra}$ in Table 4 suggest that strong relative enrichment or depletion of ^{210}Pb or intermediates, such as ^{222}Rn gas, does not occur along the flowpath.

The trends in activity ratios for the ^{235}U series are generally more regular and closer to equilibrium than for the ^{238}U series, probably because of the relatively short half-lives ranging from a fraction of a second to days for all isotopes in the series except for ^{235}U (half-life of 7.1×10^8 years), ^{231}Pa (h.l. of 3.4×10^4 years), and ^{227}Ac (h.l. of 27.7 years). Both $^{227}\text{Th}/^{227}\text{Ac}$ and $^{223}\text{Ra}/^{227}\text{Th}$ show trends towards isotopic equilibrium along the centerline. When ^{227}Th is removed from consideration by calculating $^{223}\text{Ra}/^{227}\text{Ac}$, an apparently slowly increasing trend of values towards equilibrium is also obtained. No other isotopes in this series were measured; however, if aqueous ^{235}U is close to isotopic equilibrium with ^{227}Ac and ^{227}Th , ^{235}U may comprise a few percent of total uranium but is somewhat below practical detection because of analytical accuracy of the method of subtracting ^{238}U from total uranium.

The only isotopes measured in the ^{232}Th series are ^{232}Th and ^{228}Th , although a significant presence of ^{224}Ra has been qualitatively noted in Nordic Main waters. The values for $^{228}\text{Th}/^{232}\text{Th}$ in Table 4 as well as the activities themselves yield irregular trends and the comparison of 1980 and 1981 data indicates strong temporal variations in values. It is not known whether the irregular behavior is real or an artifact of analytical error. Based on the apparently reliable comparison of measured and calculated total thorium in Table 3, ^{232}Th in 1980 for M16 should be 410 mBq/L, providing a 1980 $^{228}\text{Th}/^{232}\text{Th}$ value of 0.23. This value would give more consistency to 1980 data, but does not explain 1982 data.

In groundwater systems, Davidson and Dickson (1986) examined predicted

variations in activity ratios for radium isotopes among the decay series. For $^{226}\text{Ra}/^{223}\text{Ra}$, the value is predicted to approach 21.4 over long periods of time under the assumptions of the study. Along the centerline in Area A, $^{226}\text{Ra}/^{223}\text{Ra}$ values are 1.25 (M1), 1.32 (M20), 1.15 (M8), and 0.93 (M16), which are much less than the limiting value of 21.4 as a consequence of the short time period of 24 days represented by the centerline between M1 and M16, i.e. $x/V < 0.06$ years in the terminology of Davidson and Dickson. Furthermore, the increase in the ratio predicted along flowpaths in the simple geochemical scenario of Davidson and Dickson does not occur in Area A because of the variations in geochemical controls among the four geochemical zones between M1 and M17 (Morin and Cherry, 1986; Morin, 1988 A and B, these proceedings).

4. CONCLUSION

This paper has presented a field study of variations in several activity ratios along a groundwater flowpath of 20.8 meters in Seepage Area A of the Nordic Main uranium-tailings impoundment, Ontario. Because the groundwater requires approximately 24 days to travel this length of flowpath, observed variations during one sampling event are predominately the result of relatively fast geochemical-decay processes operating along the flowpath and are not closely related to temporal variations in the radioactive source (the tailings), which can account for variations along flowpaths in slow-moving groundwater systems. However, fluctuations in measured values at each piezometer from year to year in Area A can reflect temporal source fluctuations. Despite a higher degree of analytical error than major ions, significant variability in activity ratios along the flowpath are apparent. In the ^{238}U series, $^{234}\text{U}/^{238}\text{U}$ increases from 1.23 to 3.92 along the 20.8-meter flowpath and the other ratios generally show an initial increase or decrease followed by a decrease or increase, respectively. These trends are probably related to the four geochemically active zones that are crossed by the flowpath (Morin and Cherry, 1986). Notable isotopic disequilibrium is common in the ^{238}U series. In contrast, the measured portion of the ^{235}U series is much closer to equilibrium and steady trends towards equilibrium are apparent along the flowpath in Area A. The upper portion of the ^{232}Th series shows no clear trends along the flowpath.

This paper has also reviewed previous uranium-mining-related and non-mining-related studies. In comparison with other mining-related studies, the increase in $^{234}\text{U}/^{238}\text{U}$ along the flowpath in Seepage Area A is consistent with Veska's (1983) data (Table 2). The $^{230}\text{Th}/^{238}\text{U}$ values in Area A are less than 1 in general agreement with ratios in neutral-pH groundwater in Table 2. Also, $^{226}\text{Ra}/^{230}\text{Th}$ in Area A is significantly greater than 1 unlike other reported values except that for the Nordic Main tailings reported in Cherry et al. (1982). $^{226}\text{Ra}/^{234}\text{U}$ in Area A is close to 1 whereas the data of Veska (1983) generally decrease to values around 0.1 along the flowpath. Finally, $^{210}\text{Pb}/^{226}\text{Ra}$ in Area A is generally around 0.01-0.09, which is among the lowest values found for tailings and flowpaths near tailings (Table 2). No comparisons with previous mining-related studies can be made for the ^{232}Th and ^{235}U series.

In comparison with previous non-mining-related studies, the values in

Area A as well as ratios from Table 2 generally fall within the ranges in Table 1. This reflects the high degree of variability in non-mining-related values as a consequence of the differences in scales of time and distance among these studies.

In summary, there are similarities and differences among sites. The similarities, differences, and wide ranges of activity ratios in the ^{238}U series reflect (1) the numerous available suites of geochemical and decay controls for regulating each isotope of each element at any one site and (2) the differences in scale among the sites. At this time, there are insufficient studies to define any common trends or ranges of activities in the ^{235}U and ^{232}Th series along groundwater flowpaths.

ACKNOWLEDGEMENTS

This study was supported by: Strategic Grant G0679 from the Natural Sciences and Engineering Research Council of Canada, research agreements with CANMET (Department of Energy, Mines, and Resources Canada) and Rio Algom Limited, and a research contract from the National Uranium Tailings Program (CANMET, EMR) to Morwijk Enterprises. We wish to thank Rio Algom Limited for granting access to the Nordic Main area and Tjoe Lim (CANMET), Nand Dave (CANMET), and Al Vivuyurka (Rio Algom Ltd.) for their advice and assistance on radionuclide analyses.

REFERENCES

- CBCI Limited. 1985. A Study of Naturally Occurring, Radionuclide Bearing Deposits at Portland Creek, Newfoundland. Final Report for National Uranium Tailings Program (Canadian Department of Energy, Mines, and Resources) Contract OSQ84-00074.
- Cherry, J.A., T.A. Shepherd, and K.A. Morin. 1982. Chemical composition and geochemical behavior of contaminated groundwater at uranium tailings impoundments. SME-AIME Annual Meeting, Dallas, Texas, Feb. 14-18, Preprint No. 82-114.
- Cowart, J.B. 1981. Uranium isotopes and ^{226}Ra content in the deep groundwaters of the Tri-State region, USA. *Journal of Hydrology*, 54, p.185-193.
- Cowart, J.B. 1980. The relationship of uranium isotopes to oxidation/reduction in the Edwards Carbonate Aquifer of Texas. *Earth and Planetary Science Letters*, 48, p.277-283.
- Davidson, M.R., and B.L. Dickson. 1986. A porous model for steady state transport of radium in groundwater. *Water Resources Research*, 22, p.34-44.
- Frohlich, K., R. Gellermann, and K. Runge. 1981. On the migration of uranium isotopes in sandstone aquifers. IN: International Symposium

- on Migration in the Terrestrial Environment of Long-lived Radionuclides from the Nuclear Fuel Cycle, Knoxville, Tennessee, 27-31 July 1981, IAEA-SM-257/88P.
- Haji-Djafari, S., P.E. Antommaria, and H.L. Crouse. 1979. Attenuation of radionuclides and toxic elements by in situ soils at a uranium tailings pond in central Wyoming. IN: Zummer T.F. and Riggs C.O., editors, Permeability and Groundwater Contaminant Transport, ASTM Symposium, Philadelphia, p.221-242.
- Highland, W.R., L.T. Murdock, and E. Kemp. 1981. Design and seepage modeling studies of below-grade disposal, West Gas Hills, Wyoming. Symposium on Uranium Mill Tailings Management, Fort Collins, Colorado, Oct. 26-27, Geotechnical Engineering Program, Civil Engineering Department, Colorado State University.
- Hoffman, G.L. and S.J. Playton. 1981. Preliminary Report On The Evaluation Of The Ground-water Hydrology In The Vicinity Of Petrochemicals' Tailings Reservoir, Shirley Basin, Wyoming, by Hydro-Engineering, Casper, Wyoming.
- Kaufman, M.I., H.S. Rydell, and J.K. Osmond. 1969. $^{234}\text{U}/^{238}\text{U}$ disequilibrium as an aid to hydrologic study of the Floridan Aquifer. *Journal of Hydrology*, 9, p. 374-386.
- Kronfeld, J. 1974. Uranium deposition and Th-234 alpha-recoil: An explanation for extreme U-234/U-238 fractionation within the Trinity Aquifer. *Earth and Planetary Science Letters*, 21, p.327-330.
- Kronfeld, J., and J.A.S. Adams. 1974. Hydrologic investigations of the groundwaters of central Texas using U-234/U-238 disequilibrium. *Journal of Hydrology*, 22, p.77-88.
- Laul, J.C., M.R. Smith, and N. Hubbard. 1985. Behavior of natural uranium, thorium and radium isotopes in the Wolfcamp Brine aquifers, Palo Duro Basin, Texas. *Mat. Res. Soc. Symp. Proc. Vol. 44, Materials Research Society*, p.475-482.
- Moffett, D. and M. Tellier. 1978. Radiological investigations of an abandoned uranium tailings area. *Journal of Environmental Quality*, 7, p.310-314.
- Morin, K.A. 1988 A. Physical and chemical hydrogeology of uranium tailings in Canada and the United States of America. *International Groundwater Symposium of the International Association of Hydrogeologists*, these proceedings.
- Morin, K.A. 1988 B. Groundwater contamination from precious-metal, base-metal, uranium, and potash mining operations. *International Groundwater Symposium of the International Association of Hydrogeologists*, these proceedings.
- Morin, K.A. 1983. Prediction Of Subsurface Contaminated Transport In Acidic Seepage From Uranium Tailings Impoundments. Ph.D. Thesis, Department of Earth Sciences, University of Waterloo, Ontario.
- Morin, K.A. and J.A. Cherry. In press. Field investigation of a small-diameter, cylindrical, contaminated groundwater plume emanating from a pyritic uranium-tailings impoundment. *ASTM Special Publication ASTM-SP-963 based on the ASTM Symposium on Field Methods for Groundwater Contamination Studies and Their Standardization*, February 2-6, 1986, Cocoa Beach, Florida.
- Morin, K.A. and J.A. Cherry. 1986. Trace amounts of siderite near a uranium-tailings impoundment, Elliot Lake, Ontario, and its implication in controlling contaminant migration in a sand aquifer. *Chemical Geology*, 56, p.117-134.
- Morin, K.A. and J.A. Cherry. Submitted. Migration of acidic groundwater seepage from uranium-tailings impoundments. 3. Simulation of the conceptual model with application to Seepage Area A and other case studies.
- Morin, K.A., J.A. Cherry, N.K. Dave, T.P. Lim, and A.J. Vivyurka. Accepted, A. Migration of acidic groundwater seepage from uranium-tailings impoundments. 1. Field study and conceptual hydrogeochemical model. Accepted for publication by the *Journal of Contaminant Hydrology*.
- Morin, K.A., J.A. Cherry, N.K. Dave, T.P. Lim, and A.J. Vivyurka. Accepted, B. Migration of acidic groundwater seepage from uranium-tailings impoundments. 2. Geochemical behavior of radionuclides in groundwater. Accepted for publication by the *Journal of Contaminant Hydrology*.
- Osmond, J.K., and J.B. Cowart. 1982. Chapter 9: Groundwater. IN: M. Ivanovich and R.S. Herman, eds., *Uranium Series Disequilibrium: Applications to Environmental Problems*, Clarendon Press, Oxford.
- Osmond, J.K., and J.B. Cowart. 1974. U-234/U-238 variations in a sandstone aquifer. *EOS*, 55, p.458.
- Taylor, M.J. 1980. Radionuclide movement in seepage and its control. *Proceedings of the First International Conference on Uranium Mine Waste Disposal*, May 19-21, Vancouver, British Columbia, SME-AIME, p. 205-244.
- Taylor, M.J., and P.E. Antommaria. 1978. Immobilization of radionuclides at uranium tailings disposal sites. *Symposium on Uranium Mill Tailings Management*, Fort Collins, Colorado, Nov. 20-21, Civil Engineering Department, Colorado State University.
- Titayeva, N.A., V.A. Filonov, V. Ya. Ovchenkov, T.I. Veksler, A.V. Orlova, and A.S. Tyrina. 1973. Behavior of uranium and thorium isotopes in crystalline rocks and surface waters in a cold wet climate. *Geochemistry International*, 10, p. 1146-1151.
- Veska, E. 1983. Origin And Subsurface Migration Of Radionuclides From Waste Rock At An Abandoned Uranium Mine Near Bancroft, Ontario. Ph.D. Thesis, Department of Chemistry, University of Waterloo, Ontario.

TABLE 1
Activity Ratios At Non-Mining-Related Sites

238U DECAY SERIES							
Site	Place in flow sys.	$\frac{234Th}{238U}$	$\frac{234U}{238U}$	$\frac{230Th}{234U}$	$\frac{226Ra}{234U}$	$\frac{226Ra}{230Th}$	OTHER
Floridan Aq	gen*		0.5-1.2				
Polar Ural	gen		0.9-1.27	0.90, 0.56			
Trinity Aq	recharge 10's of km		1-2 5-12				
Carrizo Sdst	recharge 15 km >15 km		<1.0 9 approx. 2 approx				
Red Desert	updip of U barrier downdip		0.8-2.5 1.6-9.7				
Edwards Aq	gen, oxid gen, red gen, mixed		1.07-1.22 1.62-2.66 0.79-1.40				
Palo Duro Bas	gen	25-50	1.1-1.3	0.19-1.11		2-3x10 ⁴	$\frac{222Rn}{226Ra}$ 0.8-2.4
Tri-St. Reg	unmixed mixed		7-10 7-10		0.5-1.5 6-236		$\frac{210Pb}{226Ra}$ $\frac{210Po}{210Pb}$
Newfoundland	begin. 135 m		1.20 1.04	0.00273 0.0352		>1.4 0.38	10.15 26.74 0.50 0.64

(* gen = general, no clear flowpath)

232Th DECAY SERIES

Site	$\frac{228Ra}{232Th}$	$\frac{228Th}{232Th}$	$\frac{224Ra}{228Th}$
Polar Ural		7.5-17.7	
Palo Duro Bas	800-2040	7-23	90-100

235U DECAY SERIES

Site	$\frac{231Pa}{235U}$
Newfoundland	<0.04

TABLE 2
Activity Ratios At Uranium-Mining-Related Sites

Ref.-Location	$\frac{234U}{238U}$	$\frac{230Th}{238U}$	$\frac{226Ra}{230Th}$	$\frac{226Ra}{234U}$	$\frac{210Pb}{226Ra}$	OTHER
Cherry et al. (1982)						
Wyo. tailings		12.29- 23.46*	0.011- 0.058		0.17, 0.57 14.57	$\frac{210Bi}{210Pb}$ $\frac{210Po}{210Bi}$
Nordic Main "Typical"	1.00	27.78	0.01* 0.0027		12.14 1.00	1.00 1.00
Moffett & Tellier (1978)						
High-acid		4-22*	0.00058- 0.0017		2.05- 240.88	0.25-0.60
Low-acid						
Highland et al. (1981)						
Tailings "Intermed. data"		12.29 4.09*	0.012 0.001- 0.098		14.27 8.41- 8.73	$\frac{210Po}{210Pb}$ 0-0.33
Average groundwater		0.076	1.28		0.79	
Taylor (1980)						$\frac{210Po}{226Ra}$
Tailings		11.73*	0.115			0.196
WN-6S		3.73*	0.116			0.015
WN-14H		0.034*	0.0031			0.0
WN-14DA		0.068*	0.125			0.0
Veska (1983)						
L	0.97			0.33	1.15	
GR4	0.87			1.63	0.05	
GR6	0.86			1.23	0.27	
M3	1.20			0.35	0.66	
M4	1.28			0.15	<0.45	
M5	1.27			0.05	<1.00	
M6	1.57			0.08	<0.38	
M7	1.75			0.10	<0.61	
M8	1.88			0.10	<0.67	
M9	2.07			<0.65	-	

* = ²³⁸U calculated from total uranium

TABLE 3
Measured radionuclide data from the plume centerline in Area A
(values in milliBecquerels/L unless marked as molal)
(reported analytical precision +19 mBq/L)
(values >100 are rounded to the nearest 10 mBq/L)

	M1	M20	M8	M16	M17	M18	M19
Distance from M1 (m)	0	4.8	8.7	11.8	15.0	17.7	20.8
Field pH	4.40 4.24	4.61 4.60	4.90 5.18	4.98 5.68	4.86 5.43	5.65 5.21	5.50 (1980 data) 5.33 (1982)
210Pb	520 100(1982)	- 300	59 110	93 <37	37 74	- -	19 (1980) - (1981)
223Ra	3070	2670	2300	1630	-	-	- (1982)
226Ra	5930 3850	- 3520	5150 2560	2780 1520	1040 850	740 590(81)	1105(1980) 590(81) (1982)
(For 1980 Ra-226 data, M9 has an activity of 370)							
227Ac	11820	8150	7370	3960	2670	-	- (1982)
227Th	22220	15000	10000	5440	3370	-	- (1982)
228Th	130 <190(82)	- 190	190 -	93 190	<19 260	<19 -	- (1980) - (1981)
230Th	1370 1330	- 520	190 410	130 190	110 190	<19 -	1260 ⁵ (1980) - (1982)
232Th	210 <190(82)	- 37	290 190	<19 220	44 74	<19 -	74 (1980) - (1981)
Total Th(m) ¹	2.22E-7	-	3.06E-7	<2.01E-8	4.64E-8	<2.01E-8	7.81E-8 (1980)
Total Th(m) ²	2.32E-7	-	3.02E-7	4.14E-7	4.74E-8	-	7.76E-8 (1980)
234U	7570	-	4220	3040	910	-	550 (1980)
238U	6170	-	2000	930	300	-	140 (1980)
Total U(m) ³	2.07E-6	-	6.72E-7	3.12E-7	1.01E-7	-	4.73E-8 (1980)
Total U(m) ²	2.07E-6	-	6.81E-7	3.11E-7	1.01E-7	<8.40E-10	4.62E-8 (1980)
	3.57E-5	5.88E-6	3.36E-6	1.26E-6	1.26E-6	1.39E-8(81)	- (1982)
235U ⁴	46390	-	15260	6970	2260	<19	1040 (1980)

- 1 = calculated from ²³²Th
2 = measured
3 = calculated from ²³⁸U
4 = activity calculated from 5% of measured total uranium
5 = anomalous M19 value

(For conversion:
atoms/L = 1.44 (Bq/L) (half-life in seconds)
disintegrations/minute/L = 60 (Bq/L)
picoCuries/L = 27 (Bq/L)
moles/L = (half-life in seconds) (10^{-23.62}) (Bq/L)

TABLE 4
Activity ratios for the natural decay series along the plume centerline

	M1	M20	M8	M16	M17	M19
<u>238U SERIES</u> ²³⁴ U/ ²³⁸ U	1.23	-	2.11	3.27	3.03	3.92 (1980)
²³⁰ Th/ ²³⁴ U	0.18	-	0.05	0.04	0.12	2.29 ¹ (1980)
²³⁰ Th/ ²³⁸ U	0.22	-	0.11	0.13	0.36	9.00 ¹ (1980)
²²⁶ Ra/ ²³⁰ Th	4.33 2.89	- 6.77	27.11 2.90	21.38 8.00	9.45 4.47	0.09 ¹ (1980) - (1982)
²²⁶ Ra/ ²³⁴ U	0.78	-	1.22	0.91	1.14	0.20 ¹ (1980)
²¹⁰ Pb/ ²²⁶ Ra	0.09 0.03	- 0.09	0.01 0.09	0.03 <0.02	0.04 0.09	0.17 ¹ (1980) - (81/82)
²¹⁰ Pb/ ²³⁸ U	0.08	-	0.03	0.10	0.12	0.14 (1980)
<u>235U SERIES</u> ²²⁷ Th/ ²²⁷ Ac	1.88	1.84	1.36	1.37	1.26	- (1982)
²²³ Ra/ ²²⁷ Th	0.14	0.18	0.23	0.30	-	- (1982)
²²³ Ra/ ²²⁷ Ac	0.26	0.33	0.31	0.41	0.37	- (1982)
<u>232Th SERIES</u> ²²⁸ Th/ ²³² Th	0.62 -	- 5.14	0.66 -	>4.89 ² 0.86	<0.43 3.51	- (1980) - (1981)

- ¹ = activities of ²³⁰Th and ²²⁶Ra may be erroneous
² = value should be 0.23 based on calculated ²³²Th from measured total thorium in Table 3

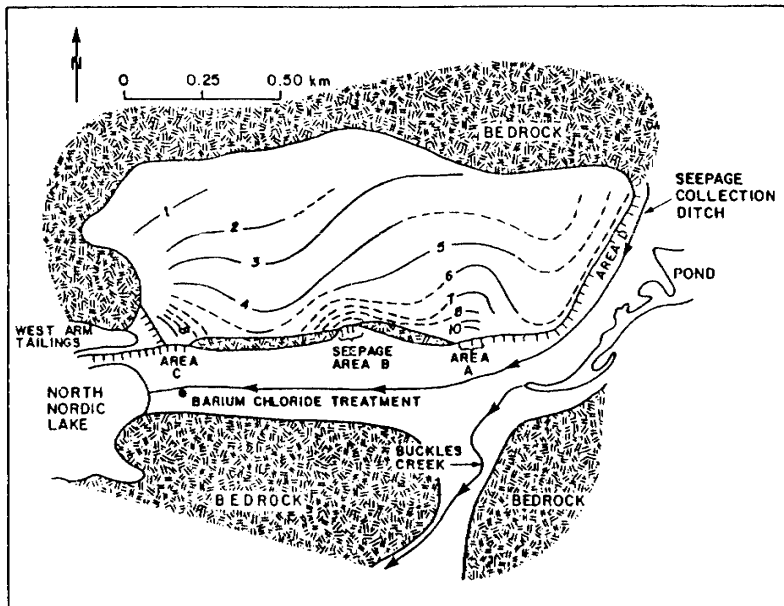


Figure 1. Nordic Main impoundment near Elliot Lake, Ontario.
Depth-to-water-table contours in the tailings in meters.

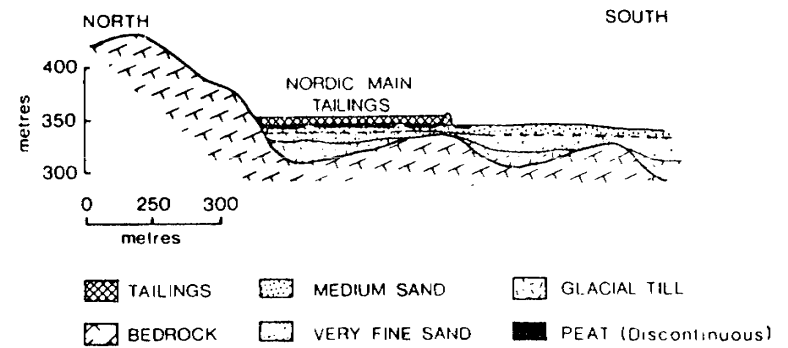


Figure 2. Generalized north-south cross-section through Seepage Area A.

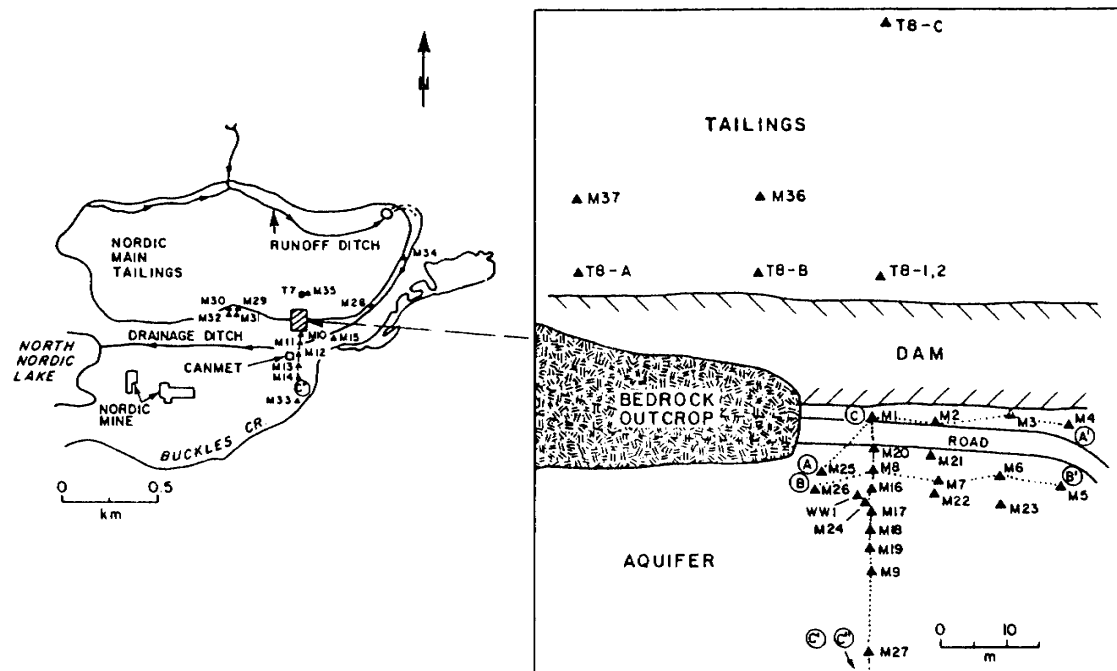


Figure 3. Locations of piezometers in the Nordic Main area.