

GEOCHEMICAL EXPLORATION FOR URANIUM IN NORTHERN LAKES

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ABSTRACT

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In the course of a decade the use of lake sediments and waters for mineral exploration in northern regions of the world has developed from infancy to a widely used and successful method. Of all of the mineral commodities that are sought, the method has been most effective for uranium exploration. This is because of the superior mobility of uranium in surface waters, that allows the element to disperse widely from its source. The effectiveness of the methods for uranium in organic-rich terrain, such as the southern Canadian Shield, has been puzzling. For in these regions the dispersion of base metals is restricted by their precipitation in swamps and organic soils. It is suggested that the good mobility of uranium in such terrain may be explained by the presence of stable, soluble organic-uranium "complexes" that allow the metal to pass through organic-rich traps.

Two possible sampling media are available for detecting uranium mineralization within the drainage basins of lakes. These are organic-rich, centre-lake sediments and surface waters. The centre-lake sediments are more suitable than nearshore sediments because of their greater consistency and because they contain a higher proportion of hydromorphically dispersed uranium. Waters have certain advantages over centre-lake sediments, such as lower sampling and preparation costs and even greater homogeneity. However, at low concentrations of uranium, the present analytical methods for water are less satisfactory than those for sediments.

The pH of lakewater has remarkably little effect on the partitioning of uranium between organic-rich sediment and water over the range 5.0–7.4. Above 7.4 there is a marked increase in the median uranium content of lake waters, but little change in the median uranium content of organic sediments. This increase in the waters with increasing alkalinity is believed to be largely at the expense of organic-poor sediments, particularly those deposited in an oxygenated nearshore environment.

In glaciated terrain, such as the Canadian Shield, the development of anomalies in lakes is a two-stage process. Firstly, uranium-rich detritus was transported down-ice from the mineralized source. Secondly, the metal is dispersed in solution from this detritus into the lakes. Because of this origin, lake anomalies are most effectively followed up by boulder tracing.

Apart from detecting mineralization within the drainage basins of lakes, attention has recently been directed to methods of detecting mineralization in the rocks beneath lakes. This is because both lakes and uranium deposits have a propensity to occur along faults.

The measurement of gases, such as helium and radon, is likely to be useful for this purpose. However, they present difficulties because of their ready exchange between surface waters and the atmosphere. The development of suitable methods for detecting mineralization beneath lakes is likely to be slowed by the difficulty of obtaining sufficient resources to carry out the three-dimensional orientation surveys necessary to trace the indicator elements from their source to the lake.

Brief case histories are given of the lake sediment and water anomalies that derive from the Key Lake, Rabbit Lake and Midwest Lake uranium deposits, Saskatchewan.

INTRODUCTION

At the start of the 1970's, the use of lake sediments and waters for mineral exploration was being investigated by a few geochemists and geologists. In the intervening years methods have been developed and refined and have been shown to be effective in outlining a variety of types of mineralization within northern shield areas of the world. Thus, at the close of the decade, most companies seeking metallic minerals in these regions sample lake sediments and/or waters as part of their exploration program.

Of the various types of metallic mineralization that have been sought using geochemical lake surveys, these methods are most notably effective for uranium mineralization. The primary reason for this is the high mobility of the element in surface waters, even in the organic-rich environments that are present in the southern Canadian Shield.

There have been a number of recent reviews that, collectively, cover most of the topics that relate to lake geochemistry in the search for uranium. These include a review of lake sediment geochemistry by Coker et al. (1979); a review of the processes responsible for the dispersion of uranium and its daughters in the surface and near-surface environment by Dyck (1979); the low-temperature geochemistry of uranium by Langmuir (1978); and a review of methods of geochemical analysis, with particular reference to uranium, by Ward and Bondar (1979).

In this paper no attempt has been made to cover again in detail the topics discussed in the above reviews. Instead, attention is directed to the data that may assist the exploration geologist in selecting the most appropriate methods and sampling media. Since uranium-organic interactions play such an important part in the dispersion and fixation of uranium, and have not been described in detail in the above papers, this topic is reviewed here.

The majority of the data described in this report come from the Canadian Shield. Similar principles may be applied to other northern regions of the world where lakes are abundant, such as Fennoscandia or Alaska. For lake sediment studies, the current status of work in these other regions has been given by Coker et al. (1979).

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LAKE SYSTEMS

In view of the great variety of limnological environments and sediments that occur, even within small areas, it is, perhaps, surprising that coherent geochemical patterns are obtained from lake samples that reflect the abundance of uranium in surface rocks and overburden. Of particular consequence is the variation in pH and in the organic and oxygen content of waters and sediments. Variation in these factors may have a marked effect on the mobility of uranium.

Lake waters may be either thermally stratified or unstratified. Unstratified waters are oxygenated. But stratified waters may be either oxygenated throughout (orthograde oxygen distribution) or show a decrease in oxygen with depth (clinograde). The depth and size of a lake determines in large part whether it will be stratified or not, and the amount of organic material and nutrients determines whether its bottom waters will be anaerobic. For northwestern Ontario, Schindler (1971) found that shallow (<4 m) lakes were unstratified and nearly saturated with oxygen; lakes of 5–12 m depth had bottom waters that tended to become anaerobic by late summer; while deeper lakes were eutrophic with anaerobic bottom waters. From the south to the north of the Canadian Shield, there is a decrease in biological activity and an increase in wind strength. These factors combine to reduce the frequency of lakes with anaerobic bottom waters in the north.

Broadly speaking, lake sediments from the Canadian Shield may be categorized as centre-lake or nearshore types (Fig. 1). The former are modern sediments, deposited in the deeper parts of the lakes since the retreat of the continental glaciers. Most nearshore material comprise older glacial and post-

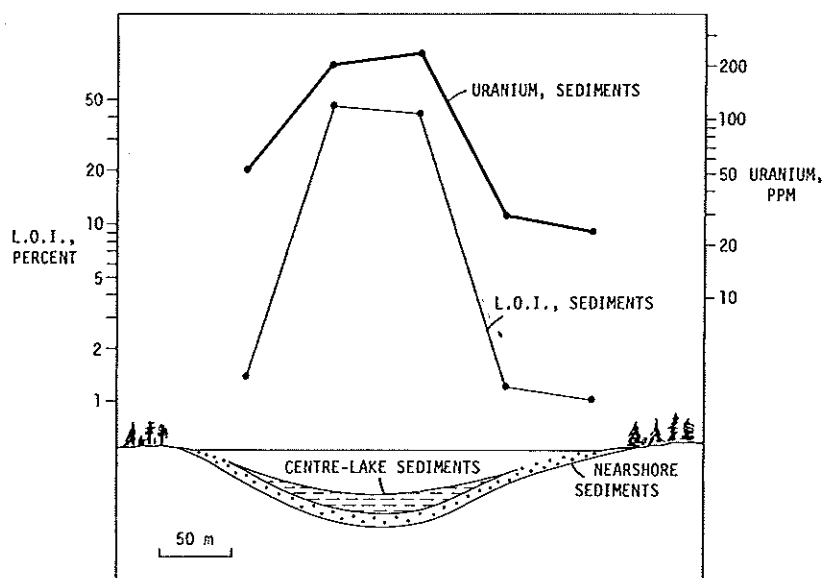


Fig. 1. Lake sediment sampling profile across Bow Lake, Ontario. (Surface sediments, 0–5 cm).

glacial sediment, such as till and outwash sand, that has been reworked by waves. Modern sediment may also occur in nearshore locations, particularly offshore from stream inflows. If a thousand samples of nearshore sediment were collected and another thousand from centre-lake, the observer would note that, on average, the centre-lake material was, finer, more homogeneous, richer in organic material, and reduced. However, he would also note many exceptions to this generalization.

In selecting the most suitable sediment for mineral exploration, the choice is between the fine-grained, organic centre-lake sediment or the coarser, predominantly inorganic nearshore sediment. Nearshore sediments have been successfully used for uranium exploration (Cameron and Allan, 1973; Minatides and Slatt, 1976). However, the centre-lake material are now generally accepted to be more suitable. Apart from being more uniform than most nearshore sediment, this material contains a higher proportion of uranium that has been precipitated from lake water. The latter is an important consideration since one is seeking labile uranium that has travelled some distance from mineralization, not the uranium contained in rock forming minerals. An additional advantage of centre-lake sediments is that they may be sampled more rapidly by helicopter than nearshore sediments. In the south of the Shield, trees may prevent a helicopter nearing the shore and in the north the nearshore sediments are often frozen. Nearshore sediments require sieving to recover the fine fraction, but this is not usually required for the centre-lake samples.

The contrast between nearshore and centre-lake sediment is demonstrated by a profile (Fig. 1) across Bow Lake, Ontario. Note the much higher levels of uranium fixed in the centre-lake sediments. This is because of a number of circumstances: higher organic content (expressed as loss on ignition, L.O.I.) to fix uranium; greater abundance of clays and other fine-grained adsorbents in the centre-lake sediment; and a reducing environment on the centre-lake bottom, compared to an oxidizing one nearshore. Figure 2 shows that the

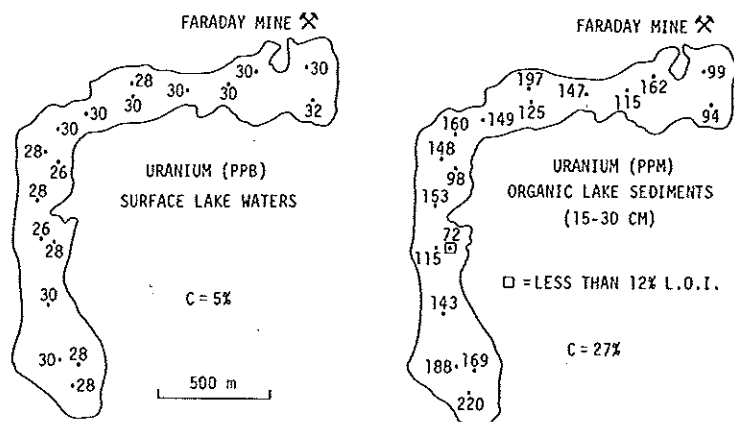


Fig. 2. Distribution of uranium in surface lake waters and centre-lake, organic sediments, Bow Lake, Ontario. (Subsurface sediments, 15–30 cm). C = coefficient of variation.

distribution of uranium in the centre-lake sediments of this lake is reasonably consistent. The lake would have been identified as being strongly anomalous in this element irrespective of the location of the sampling site. It is immediately adjacent to the Faraday uranium mine in the Bancroft district of Ontario. These sediment samples were collected from below the level affected by contamination from the mine. This illustration also shows that the homogeneity of the sediment is far less than that of the well mixed surface water. The data used to prepare Figs. 1 and 2 were obtained from Coker (in prep.).

HYDROMORPHIC DISPERSION OF URANIUM

The basis for the measurement of uranium in lakes as a guide to deposits is its high mobility in oxygenated surface waters. In the northern Shield areas of the world, where lakes are so abundant, relief is generally low. Thus rock and mineral detritus can only be transported short distances down-drainage. However, oxygenated waters dissolve many uranium-bearing minerals and may transport the metal considerable distances before it is precipitated.

The essentially hydromorphic nature of uranium dispersion is most readily apparent in the uranium anomalies present in surface waters from the vicinity of deposits. Examples of this will be described later in this report. Other evidence has been provided by Dyck (1974) and Levinson et al. (1978) who have shown that the uranium present in lake sediments from uraniferous districts of Saskatchewan is considerably out of equilibrium with its daughter products. The thorium, radium and lead produced by the decay of ^{238}U and ^{234}U are relatively immobile and are retained in the rocks, in the soils, and in drainage sediments proximal to the uraniferous source. Their data suggest that, apart from the background levels of the element present in the silicate and other inorganic component of the sediment, most of the uranium present in centre-lake, organic sediments has been transported in solution.

In an oxygenated environment, uranium is most readily soluble in acidic, or in moderately alkaline to alkaline waters. The solubility of uranium in alkaline waters is due to the formation of uranyl di- and tri- carbonate and penta-hydroxy complexes. In typical northern surface waters the amount of dissolved uranium is likely limited, not by the solubility of any particular uraniferous mineral, but rather by its adsorption on various organic and inorganic compounds present in sediments or in suspension. The pH range of minimum solubility for uranium minerals, weakly acidic to neutral water, is also the range for maximum sorption of uranyl ion on inorganic adsorbents, such as ferric oxyhydroxide (Langmuir, 1978) or on organic materials such as humic material (Fig. 3).

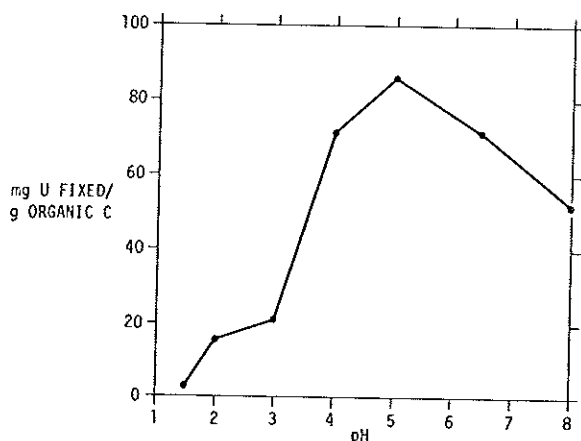


Fig. 3. Effect of pH on the adsorption of uranium by organic material (after Bloomfield and Kelso, 1973).

INFLUENCE OF ORGANIC MATTER ON URANIUM

The affinity between uranium and organic matter is one of the strongest of all the metals. Pauli (1975) showed that a greater quantity of UO_2^{2+} was bound to "aqua humus" than either Cu^{2+} , Zn^{2+} , Ni^{2+} , Pb^{2+} , or Cd^{2+} . This affinity is generally considered to result in the fixing of uranium on organic solids. However, it is most important to emphasize that soluble organic-uranium compounds are also formed. Humic and fulvic acids have a strong solubilizing effect on sulphides, oxides, silicates and other minerals (Bondarenko, 1968; Baker, 1973; Singer and Navrot, 1976). Bloomfield and Kelso (1973) found that decomposing organic matter increased the solubility of UO_2 by a factor of >140 at pH 8.

The exact nature of soluble organic-uranium associations is not clear. It is often considered that organic-metal complexes are formed. But Stumm and Bilinski (1972) suggest that the major role played by dissolved humic material is to form protective colloids that stabilize simple inorganic metal complexes. Whatever their nature, these soluble organic-uranium associations are not stable with time. Bloomfield and Kelso (1973) showed experimentally that, over a period of months, there was a substantial loss of filterable uranium associated with organic matter of molecular weight <1000 . There appeared to have been coagulation of the humic material of low molecular weight and its associated uranium into solids of high molecular weight.

A question of interest is whether soluble organic matter can delay the fixing of uranyl ion in anaerobic bottom water, sediment, or swamp. It is known that the content of certain heavy metals in the pore waters from sulphide-bearing sediment may be substantially greater than the amounts predicted from calculations of sulphide solubility (Elderfield and Hepworth, 1975). This may, in part at least, be caused by the formation of stable metal-organic complexes (Presley et al., 1972; Hallberg, 1974). In the case of U^{4+} ,

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it appears that soluble organic complexes may exist (Jackson et al., 1978).

Fixation of uranyl ion by organic matter can be accomplished in two principal ways. The first is the adsorption of the ion by organic solids. This process may initially involve a reduction step (Szalay and Szilagyi, 1968). The second is the reduction of UO_2^{2+} to insoluble UO_2 by H_2S and other reductants in the anaerobic environments created by an abundance of organic material.

There is empirical evidence to suggest that, moving from the north to the south of the Canadian Shield, lake sediment and water sampling become less effective in exploration for base metals, e.g. copper or zinc. Presumably, this is because these metals are more readily fixed by the abundant organic matter of the southern Shield and disperse less widely from mineralization. In many cases, base metals released in solution from mineralization are precipitated in swamps before they enter the lakes. This has been demonstrated by unpublished studies in the Grenville Province, Ontario (E.W. Boyle and J. Nichol, pers. commun., 1977–1980). For uranium the same does not appear to hold. Lake geochemical methods for uranium exploration appear to be of comparable effectiveness throughout the Shield. It is suggested here that this very important difference between uranium and base metals is due to the formation of soluble uranium complexes with organic material which allows uranium to pass through organic-rich traps that might precipitate other metals.

It is difficult to provide a quantitative basis for the above remarks, because of the lack of comparative studies on the dispersion of elements in different surficial environments. Also, data on the relative amounts of soluble organic matter in lakes from different regions are lacking. In this respect the recent development of laser-induced fluorimetry (L.I.F.) for the analysis of uranium in water (Robbins, 1978) is of interest. The method is based on the fluorescence of UO_2^{2+} . Organic materials interfere with the determination of uranium in three principal ways (Hall, 1979; G.E.M. Hall, pers. commun., 1980). Firstly, humic acid may quench the fluorescence of UO_2^{2+} (deactivation by collision). Secondly, certain organic compounds also fluoresce. Both these forms of interference can be corrected and in doing so the analyst is provided with a rough estimate of the amount of organics present. In the course of analysing surface waters from four regions of Canada, Hall (1979) found moderate to severe interference in samples from three forested regions. But samples from northern permafrost terrain (Melville Peninsula) showed nil to slight interference. These estimates have been confirmed by recent analyses of organic carbon in the waters. Five samples from Melville Peninsula averaged 2.0 ppm C, while eight from north-western Ontario averaged 8.7 ppm C.

It is the third form of interference that is of the greatest relevance to this discussion. Some waters, which need not necessarily indicate a high organic loading, give much lower results by L.I.F. than by classical fluorimetry (the latter method is described by Smith and Lynch, 1969). For samples

collected in southern British Columbia and in Ontario, G.E.M. Hall (pers. commun., 1980) found the uranium content by L.I.F. to be only 10–70% of the uranium measured by classical fluorimetry. Preliminary results indicate that the uranium “lost” by L.I.F. is returned only slowly and incompletely by the type of chemical attack, such as acidification, likely to free the element from inorganic complexes. However, the metal is released by irradiating the waters with ultra-violet light. These results suggest that for those regions of Canada with high rates of organic growth and decomposition, substantial amounts of uranium may be carried in solution as stable organic complexes. The laser fluorimeter appears to be a powerful tool for investigating the distribution of these complexes.

INFORMATION PROVIDED BY NATIONAL GEOCHEMICAL RECONNAISSANCE (N.G.R.) SURVEYS

Many tens of thousands of lake sediment and water samples have been collected in the course of N.G.R. surveys (Hornbrook and Garrett, 1976; Cameron and Hornbrook, 1976; Coker et al., 1979) carried out in support of the Uranium Reconnaissance Program (Darnley et al., 1975). These provide a unique opportunity to examine the relation of organic matter, pH, and other variables to the uranium content of sediments and waters.

The areas sampled by lake surveys to 1978 are shown in Fig. 4. Data from most of these areas have been examined in preparing this paper. The conclusions obtained are broadly similar for all areas, so that the data illustrated are mainly from one area only — northwestern Ontario. This is typical Shield terrain with heavy forest cover. It should be noted that the chemistry of some of the lakes in this region have been influenced by “acid rain” (Coker and Shilts, 1979).

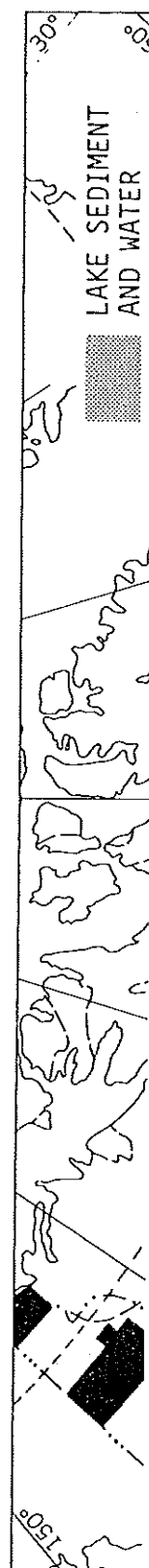
Uranium in sediment data were obtained by delayed neutron counting and uranium in water data by fission - track analysis. Iron and manganese in sediments were analyzed by atomic-absorption spectrometry following extraction with 4 M HNO₃/1 M HCl.

The metal data have been plotted as the medians for different classes of loss on ignition (L.O.I.) or pH. The median is used because it is a particularly robust measure of central tendency that is less affected by outliers than either the arithmetic or geometric means. In addition, it allows the uranium in water data sets, that are truncated by the detection limit, to be treated.

Metals versus L.O.I.

Figure 5 shows uranium, iron and manganese versus L.O.I. for 4268 centre-lake sediments from northwestern Ontario. The latter two elements are shown in addition to uranium since, when present as oxides, they are effective scavengers of trace metals.

The curve representing uranium has minima of ~1.5 ppm U in the ranges 1–10% and 80–90% L.O.I. and a maxima of 2.8 ppm over the range



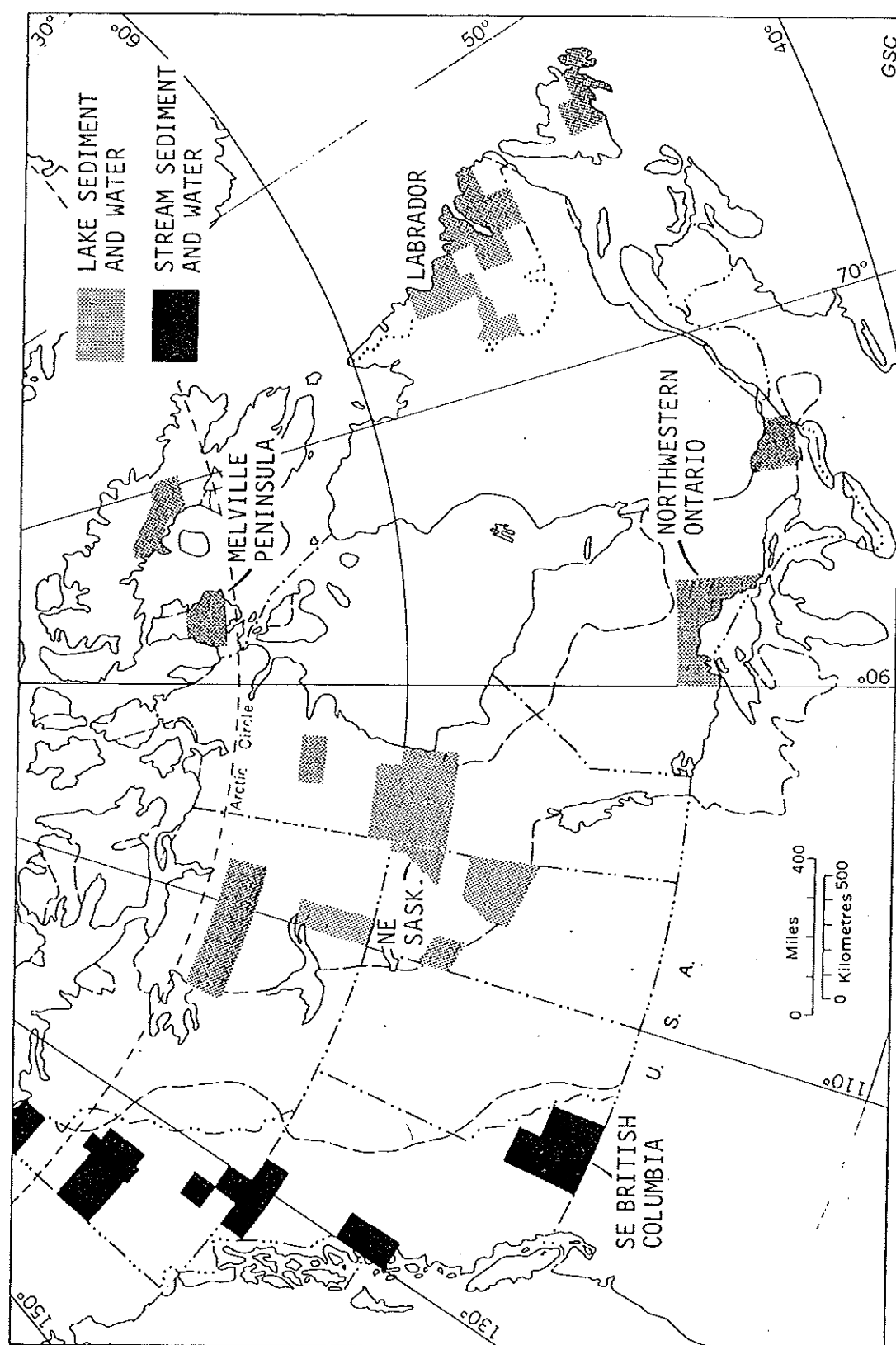


Fig. 4. Index map to National Geochemical Reconnaissance surveys.

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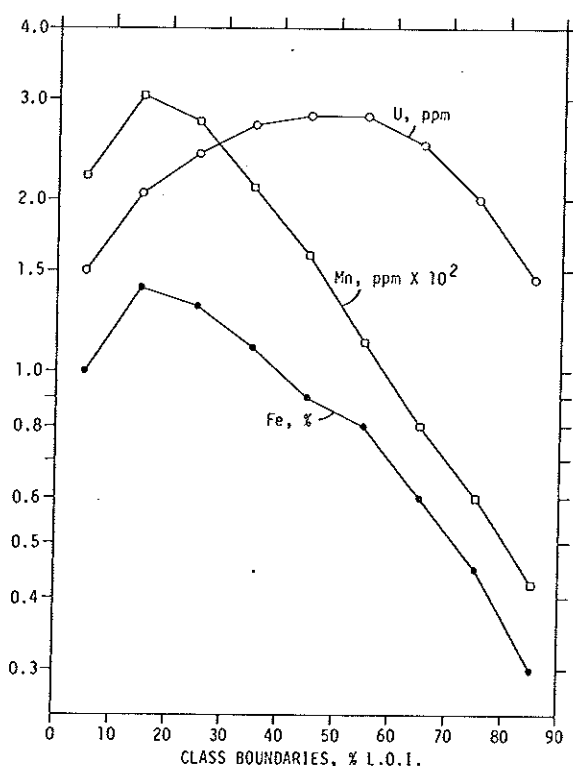


Fig. 5. Median contents of uranium, manganese and iron for classes of loss on ignition, centre lake sediments, northwestern Ontario, 4268 samples.

40–60% L.O.I. Garrett and Hornbrook (1976) were the first to show a similar, but not identical, distribution of metal (Zn) versus L.O.I. for a large group of centre-lake sediments. They attributed the strong correlation between metal and L.O.I. at low L.O.I. contents to metal adsorption on organic material. But over the middle or plateau region there is an excess of organic adsorbent, so that the amount of metal incorporated in the sediment is determined by its availability. Clearly, this is the best type of sediment to sample for exploration purposes. Finally, at high levels of L.O.I., the organics act as a diluent of the metal.

In Table I the uranium versus L.O.I. relationship is compared for northwestern Ontario, Saskatchewan and the Melville Peninsula. These may be considered to represent, respectively, southern, mid- and northern Canadian Shield terrain. Only the Melville Peninsula is north of the treeline and the effect of this on the organic content of the sediments is readily apparent. It should be noted that it is in northwestern Ontario that low-organic samples show the greatest diminution in uranium content relative to the more common L.O.I. classes.

It should be recognized that the L.O.I. term is not simply reflecting a change in organic content, but also in a number of correlated variables. These variables, although not measured, may also have influenced the metal content of the sediment. Apart from the clays and other hydrated minerals that may directly contribute to percent L.O.I., there is a correlation between L.O.I. and the depositional environment and even the local terrain. For

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TABLE I

Median uranium content of L.O.I. classes, centre-lake sediments, Canadian Shield

	Northwestern Ontario		Saskatchewan		Melville Peninsula	
Number of samples	4071		1996		2126	
Highest class median ppm U	2.8		6.1		21	
Classes of percent L.O.I.	Percent of median U to highest class median	Percent of samples in class	Percent of median U to highest class median	Percent of samples in class	Percent of median U to highest class median	Percent of samples in class
0-10	54	5	66	6	77	48
10-20	73	6	97	9	95	39
20-30	86	9	100	16	100	8
30-40	96	18	100	26	98	5
40-50	100	26	97	21	—	—
50-60	100	21	94	12	—	—
60-70	89	10	73	6	—	—
70-80	71	4	61	3	—	—

instance, centre-lake sediments with low L.O.I. tend to occur more frequently in aerobic environments. At the other end of the range, samples with >50% L.O.I. are more common in low, swampy terrain. In such areas metals are less easily mobilized from the underlying rock and overburden. Another example of "hidden" variables associated with the term L.O.I. became apparent when examining the Labrador and Ontario data sets. For the Labrador data, comprising 9046 samples, the highest median uranium contents are in the 0-20% L.O.I. classes and median uranium declines with increasing organic content. This region is close to the northern limits of forest cover and it is presumed that the uraniferous districts are, on average, less productive of vegetation because their distribution is biased towards northern locations. It is also possible that some lithologies in which uranium is more abundant, such as granites, provide a thinner and less nutritious soil. For the northwestern Ontario data set the distribution of <10% L.O.I. samples is strongly biased in favour of lakes with surface water pH in the highest range (7.5-8.2). Some 53% of low L.O.I. samples occur in these lakes although surface waters in this pH range comprise only 21% of the total. In the lakes with pH >7.5 the partitioning ratio uranium in water/uranium in sediment (U_w/U_s) is likely to be greater than for lakes with pH

<7.5. This effect may depress the uranium content of the low-organic group of sediments to a greater degree than simply the organic factor alone. It is presumed that some geological condition has produced both a lowering of sediment organic content and an increase in water pH within certain lakes. In this region relatively alkaline surface waters occur in areas underlain by calcareous drift or by carbonate members of the Sibley Group, or carbonate iron formation (Coker and Shilts, 1979).

The above points are specifically made to point out the possible danger of "correcting" or normalizing uranium in sediment data to constant L.O.I. The curves used to normalize the data may not entirely depend on the uranium-organic relationship. The normalization of data on the basis of relationships determined in a different area or geological terrane is particularly imprudent.

Iron and manganese have a quite different relationship to L.O.I. (Fig. 5). These elements show maxima in the range 10–20%, then fall off rapidly as the sediments increase in organic matter. The rate of decrease is consistent with dilution of iron- and manganese-bearing minerals by organics. However, it is also possible that some of these metals became reduced to Fe^{2+} and Mn^{2+} when buried with the organic-rich sediment. Once in solution, these will tend to be precipitated near the boundary between aerobic and anaerobic conditions on the lake bottom. Sediments with L.O.I. contents in the range 10–20%, which show maxima for iron and manganese, may well represent these boundary conditions. The curves shown in Fig. 5 suggest that these two elements have little influence on the distribution of uranium in these lake sediments.

Uranium and pH

The greater solubility of uranium in alkaline waters can present difficulties in interpreting geochemical survey data. With increasing alkalinity, the partitioning of uranium between sediment and water changes. Surveys in areas with moderate or strong variation in alkalinity will show variation in the uranium content of its sediments and waters due to this cause (Maurice, 1977; Coker and Closs, 1979). Since it is the objective of these surveys to portray only variation correlated with the spatial distribution of uranium in rock and overburden, it is desirable to choose a sampling medium that is least responsive to varying alkalinity. Further, it may be necessary to compensate for changes in uranium content that are caused solely by alkalinity.

Figure 6 shows that pH-related changes in the partitioning of uranium between sediment and water are not trivial. The plot was obtained by computing U_w/U_s for each of 4264 Ontario sites, then obtaining median ratios for each of eight classes of pH between 5.0 and 8.2. Medians have been obtained for each of two site categories: those having sediment L.O.I. <10% and those with L.O.I. ≥10%. For the organic-rich category the ratio is remarkably similar over the pH range 5.0–7.4, but shows a sharp increase

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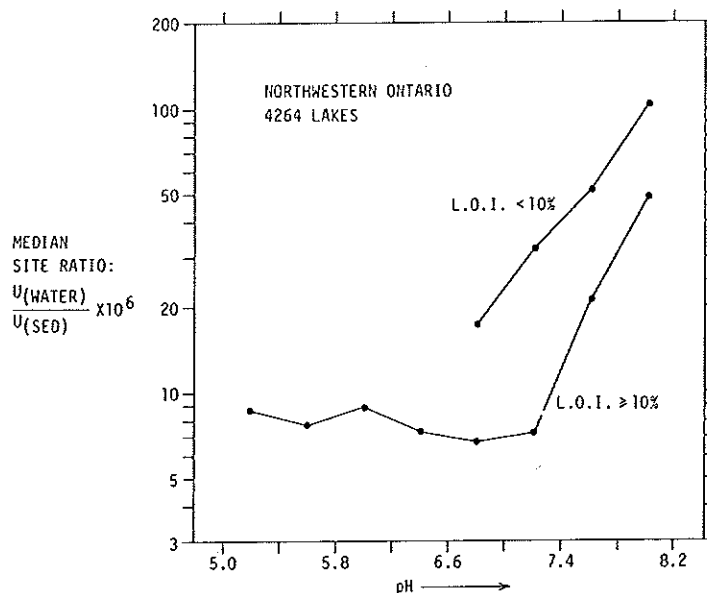


Fig. 6. Median site ratio, uranium in water to uranium in centre-lake sediment for different classes of pH. Lake sites in northwestern Ontario.

at higher pH. The low-organic sites have a higher U_w/U_s at given pH, indicative of the lesser affinity of uranium for this sediment.

In Figure 7 the ratio U_w/U_s for 6424 stream sites from southern British Columbia is superimposed on the Ontario lake site data previously shown in Fig. 6. Note the remarkable similarity in the partitioning of uranium for the two environments. The increase in U_w/U_s takes place at a lower pH for the stream sites than for the organic lake sediment sites. Although L.O.I. was not

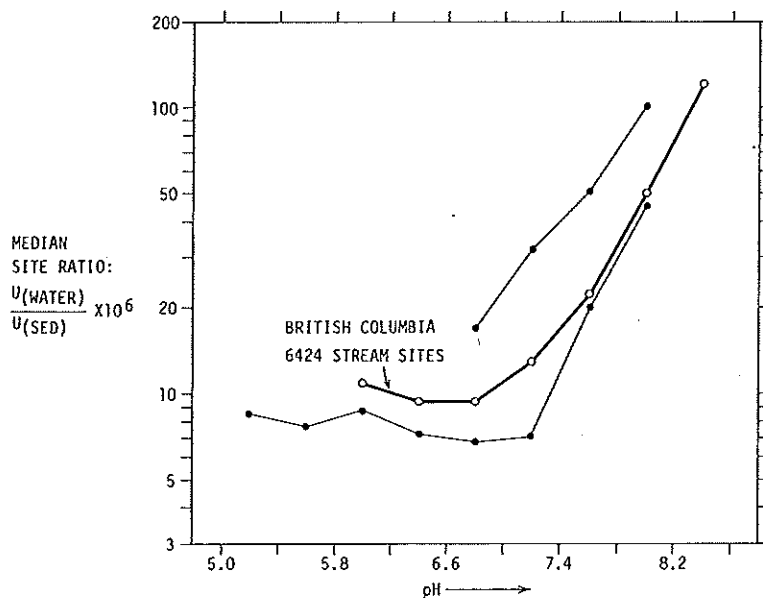


Fig. 7. Median site ratio, uranium in water to uranium in stream sediment, southeastern British Columbia. Superimposed on lake site data given in Figure 6.

measured on the stream sediment samples, they almost certainly have less organic material than the $\geq 10\%$ L.O.I. lake samples. The distribution of median U_w/U_s versus pH is broadly similar to Fig. 6 for all Shield areas examined, except Melville Peninsula, which shows a continuous increase in ratio from pH 5.9.

Figure 8 indicates that the rapid change in partitioning above pH 7.5 for the northwestern Ontario lakes is not caused by change in the uranium content of the organic-rich centre-lake sediments, but rather by a rapid increase in the median uranium content of the waters. The low-organic centre-lake sediments do show some decrease in uranium with increasing pH, but this is slight compared to stream sediments (Fig. 9). The interpretation

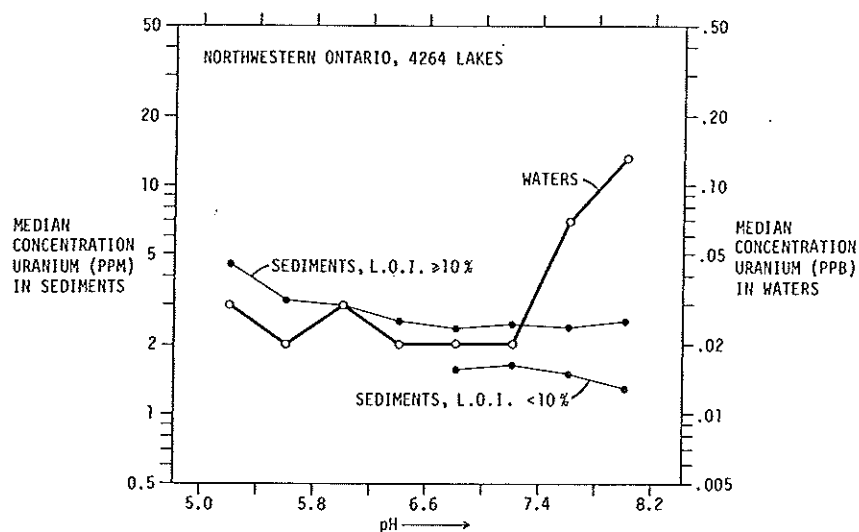


Fig. 8. Median concentration of uranium in water and uranium in sediment for classes of pH. Lake sites in northwestern Ontario.

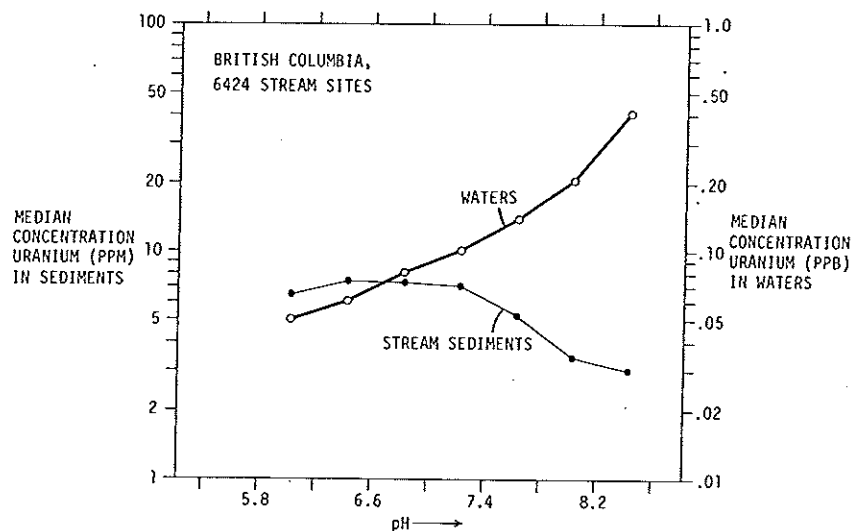


Fig. 9. Median concentration of uranium in water and uranium in sediment for classes of pH. Stream sites in southeastern British Columbia.

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placed on these data is that the affinity of uranium for organic matter plus the tendency for this element to be fixed as quadrivalent uranium in anaerobic environments is sufficient to maintain the flux of metal precipitated in $\geq 10\%$ L.O.I. sediments, even under the moderately alkaline conditions present in some Canadian Shield lakes. The flat nature of the $U_s(\geq 10\% \text{ L.O.I.})$ curve (Fig. 8) up to pH 8.2 is at variance with some experimental data which show a rapid decline in uranium adsorbed by peat over the pH range 7.5–8.2 (Lopatkina, 1967). However, other experimental data (Fig. 3 and Doi et al., 1975) show good adsorption up to the pH limits generally found in Shield lakes.

The increased uranium content of alkaline water must, obviously, be at the expense of some type of lake sediment. It is suggested that this is likely to be the nearshore, low-organic, well oxygenated sediment. For centre-lake sediment the partitioning ratio ($U_s(\geq 10\% \text{ L.O.I.})/U_s(> 10 \text{ L.O.I.})$) increases only moderately with increasing alkalinity. For the data given in Fig. 8 this ratio is ~ 1.5 for pH classes < 7.4 and ~ 1.9 for the class pH 7.5–8.2. The ratio for the Bow Lake traverse (Fig. 1) is much greater at 6.5. Since the surface waters of this lake are only moderately alkaline at pH 7.4 (Coker, in prep.), this high ratio is mainly attributable to contrasting organic centre-lake sediments with nearshore low-organic sediments. While data to show the distribution of uranium in nearshore sediments versus pH is not available, it is probably similar to that for the stream sediments shown in Fig. 9; that is, uranium decreases much more markedly with increasing pH than for centre-lake sediments.

It is clear from the above that organic-rich centre-lake sediment is a suitable sampling medium for uranium measurement, even in the moderately alkaline lakes present in the Canadian Shield. These lakes occur within carbonate terrane. The uranium chemistry of lake waters from this region is reasonably stable over the pH range 5.0–7.4 (Fig. 6). If waters of higher pH are used for exploration, it is essential that the alkalinity of the water be factored in to the interpretation. For neutral to alkaline surface waters of the composition found on the Shield there is a good correlation between alkalinity, bicarbonate content, pH and conductivity. Since the latter two measurements are more easily made than the former two, they will normally be used to help interpret uranium in water data.

Equilibrium relationship between uranium in water and in sediment

Since the dispersion of uranium in lake drainage systems is essentially hydromorphic, there is likely to be an equilibrium established between uranium present in the sediment and the water of a given lake. For north-western Ontario the uranium in water data have been separated into classes corresponding to the amount of uranium present in sediment taken from the same lake. Then for each class, medians for U_w have been computed for two categories of pH: ≥ 7.5 and < 7.5 . The resulting curves given in Fig. 10

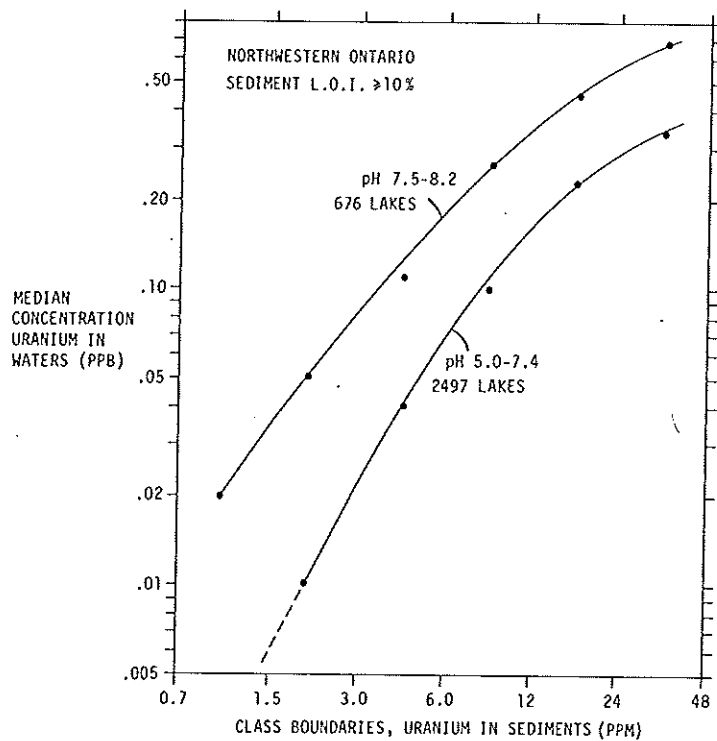


Fig. 10. Median concentration of uranium in surface lake water for classes of uranium in centre-lake sediment, northwestern Ontario lake sites.

indicate that there is a general equilibrium between uranium present in sediment and in the overlying water and that this equilibrium depends on pH. As might be expected, the raw data show a good deal of scatter about each curve reflecting the local variation in the environment that will influence the equilibrium attained within any given lake.

Both curves approximate a 45° slope over their middle to upper portions. The curves are steeper towards their base, presumably because there is a certain amount of detrital uranium present in the sediments that does not enter into the equilibrium relationship. If the pH < 7.5 curve is plotted on linear rather than logarithmic paper, it intersects the uranium in sediment axis at ~ 1.3 ppm U. This value may be considered a measure of the average amount of detrital uranium in the lake sediment of the area. Extrapolation errors are too great to allow a similar estimate to be made for the pH ≥ 7.5 curve.

SAMPLING AND ANALYTICAL CONSIDERATIONS

The distribution of dissolved metals within the surface waters of small to moderately sized northern lakes ($< 2 \text{ km}^2$) has been shown to be remarkably homogeneous. This is apparent for uranium in the data shown in Fig. 2 and, for base metals in the results reported by Cameron (1978). Regarding seasonal variation, MacDonald (1969), working in the Beaverlodge area, found

no significant differences during spring and summer (1978) for the Shield. Data were sampled from 36 samples attributed to the same agreement in sampling obtained from waters from August, 1978.

As was noted, sediment concentrations in waters. For example, homogeneity at 5% of the total apart. As noted by Garre, variance in cells and for $< 3\%$ variance in investigation (press).

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no significant variation in the uranium content of 13 control lakes, except during spring break-up. Essentially similar results were reported by Cameron (1978) for base metals in lakes from the northern part of the Canadian Shield. Dyck and Tan (1978) compared the uranium content of lake waters sampled during winter (March) and again after break-up (June). On average, 36 sampling sites had 34% less uranium in June than in March. Dyck attributes this to dilution by snowmelt. These three investigations are in agreement that seasonal variation is slight and is no impediment to the sampling of waters for mineral exploration. Quite different results were obtained by Meyer (1969) who found a 2 to 4 times more uranium in lake waters from Labrador sampling in September, 1966, compared with a July--August, 1967 sampling.

As was shown in Fig. 2, the variation of uranium in the centre-lake sediments of a lake is substantially greater than its variation in the lake waters. But even so the distribution in these organic-rich sediments is reasonably homogeneous. During N.G.R. surveys duplicate samples were collected at 5% of the sample sites. They are considered to have been collected 100 ft. apart. An analysis of variance study utilizing these data has been reported by Garrett and Goss (1979). The authors show that within-site and analytical variance is very small compared to variation between lakes within 13 km² cells and between cells. Together, the analytical and site variation account for <3% of the total variation. It should be emphasized that the within-site variance is not a meaningful estimate of the within-lake variance. An investigation of the latter has recently been carried out by Hornbrook (in press).

In deciding whether to utilize lake waters or lake sediments, the analytical precision and detection limits for uranium in the two media are an important consideration. In Fig. 11, long-term precision data (J.J. Lynch, pers. commun., 1980) have been superimposed on the uranium in water versus uranium in sediment curve given in Fig. 10 for waters of pH 5.0--7.4. This allows an estimate to be made of the relative analytical precision for the waters or sediments of any given lake. It is immediately apparent that uranium in water measurements are less precise and may be subject to constraints prescribed by the detection limits. For lakes that are moderately to strongly anomalous in uranium, this poses no serious handicap, since adequate precision is available with existing methods of water analysis. But for a weak anomaly in an area of low background, there is a difficulty. Later in this report case histories will be given of geochemical responses to uranium mineralization present in the Athabasca Sandstone. In this region typical background values are <2 ppm U in sediment and ~0.01 ppb U in waters. Thus anomalies of, say, 4 ppm and 0.02 ppb may be significant. Present analytical technology is adequate for 4 ppm U or less in sediment, but not for 0.02 ppb U and less in water.

The precision data given in Fig. 11 understate the superiority of uranium in sediment analyses. At the parts per trillion level, uranium in water

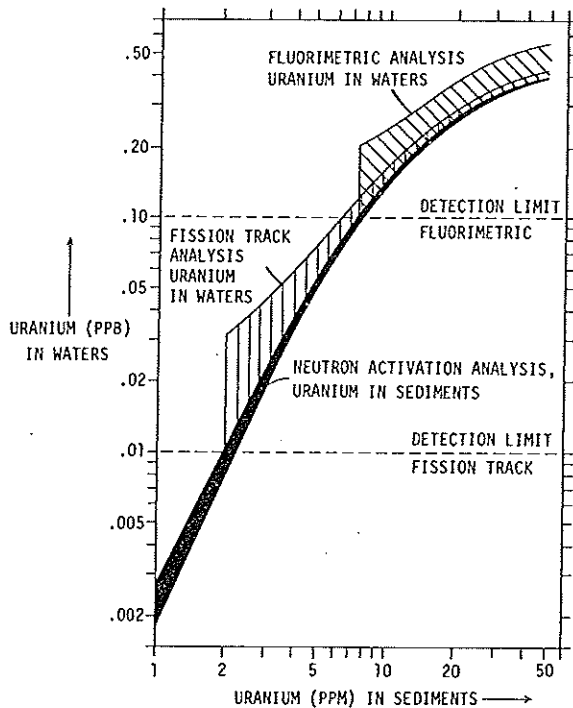


Fig. 11. One standard deviation analytical precision bands for uranium in organic lake sediments and uranium in waters. Plotted on either side of pH 5.0—7.4 curve shown on Fig. 10.

analyses requires superior technical skills, and a high level of attention to quality control. The number of sample batches that must be reanalysed for uranium in water are much greater than for sediments.

One further cause for concern in the sampling of waters for uranium exploration has been the stability of the uranium in solution over the period between sampling and analyses. Hall (1979) has described a comprehensive study where surface waters from a variety of geological environments across Canada were collected and analyzed. In addition to the stability of uranium in untreated waters, the effects of filtration and different preservatives were tested. It was shown that uranium was stable in all of the untreated waters tested and that filtration or the addition of preservatives is unnecessary.

EXAMPLES OF ANOMALIES ASSOCIATED WITH URANIUM DEPOSITS

In the section below three uranium deposits having associated lake sediment and water anomalies will be described. All three deposits are in Saskatchewan and lie near the unconformity of the Athabasca Sandstone of Helikian age on older Precambrian rocks. This region is currently the scene of the most intense exploration effort for uranium in north America. It will, undoubtedly, be the forcing ground for even more sophisticated exploration techniques, including those utilizing lake geochemistry.

In the preceeding parts of this paper emphasis has been placed on the

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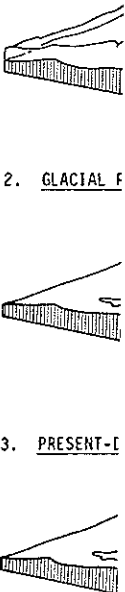


Fig. 12.

hydromorphic nature of uranium dispersion. The reader should not assume, however, that in glaciated terrain anomalies will be arranged immediately down-drainage from mineralized bedrock. Instead, the ice sheets that covered the Canadian Shield eroded mineralization and displaced the detritus down-ice. It is the weathering of this material and subsequent hydromorphic dispersion that has produced many present-day lake anomalies. Fig. 12 illustrates this process and good examples are provided by anomalies that lie down-ice from the Rabbit Lake and associated deposits (Fig. 13a, b) and from the Key Lake deposit (Figs. 14 and 15). Glacial dispersion in this region was to the southwest.

At Rabbit Lake mineralization lies close to the surface. But at Key Lake the two orebodies lie at depths of 90 m and 180 m beneath sand and, in places, beneath unmineralized Athabasca Sandstone. In this case the derivation of glacially-eroded material is not so obvious. However, detailed exploration by Uranerz Ltd. has shown that glacial till containing mineralized boulders lies directly on part of the Gartner orebody (Fig. 16). This portion of the deposit was eroded by the glacier, then covered by a thick blanket of outwash sand after the last glacial retreat. In both of these areas most lake waters have a $\text{pH} < 7.5$. The samples, therefore, relate to the horizontal portion of the U_w/U_s versus pH curve (Fig. 6) and the water and sediment anomalies correlate very well.

The data given in Figs. 14 and 15 for the Key Lake area are derived from

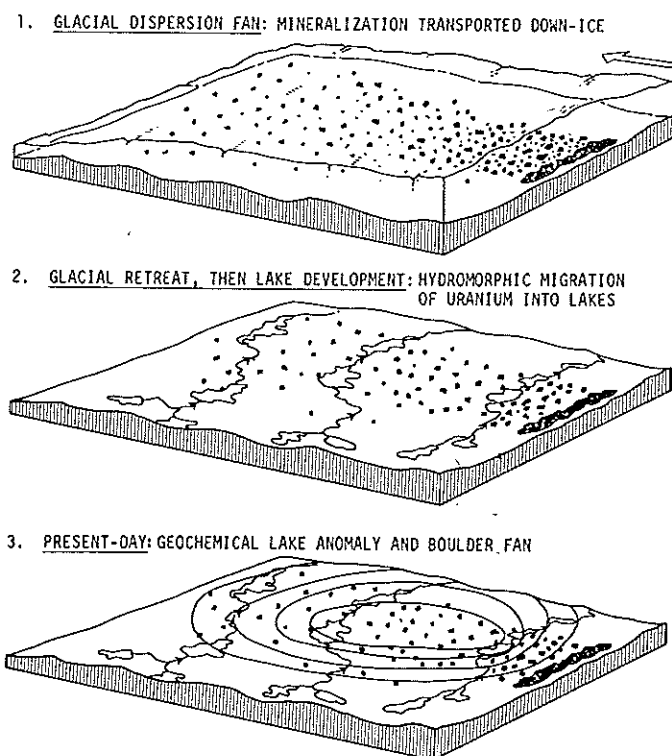


Fig. 12. Diagram illustrating the two-stage origin of lake anomalies in glaciated terrain.

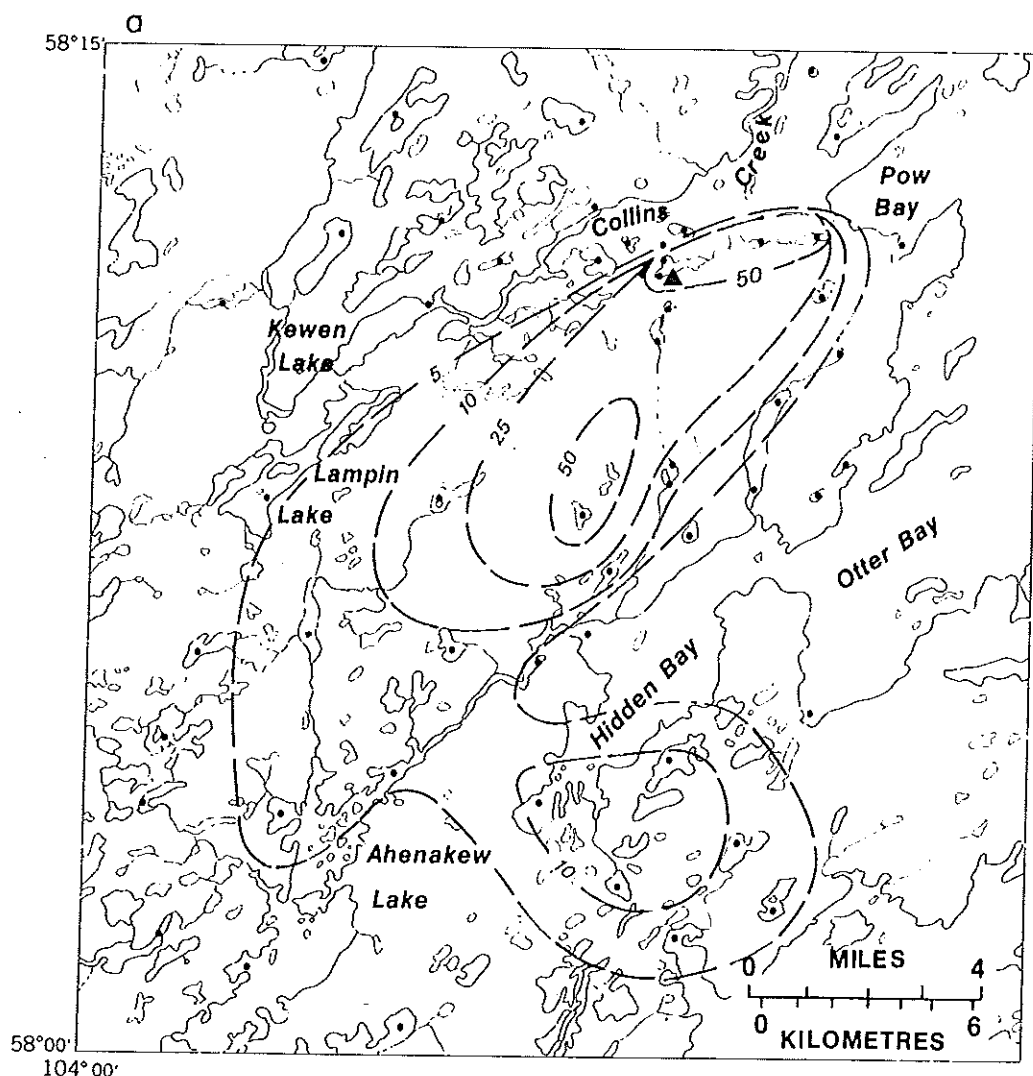


Fig. 13a. Distribution of uranium in centre-lake sediments as ppm, Rabbit Lake area, Saskatchewan. Rabbit Lake uranium deposit shown as solid triangle. (From Cameron and Ballantyne, 1977).

recent surveys by the Geological Survey of Canada (W. Dyck and Y.T. Maurice, pers. commun., 1979). Earlier lake geochemical surveys contributed substantially to the discovery of the deposit. This early work, described by Tan (1977) recognized the value of organic-rich sediments and the derivation of the lake anomaly from glacially-transported ore boulders. It is appropriate to quote here the abstract given by Tan: "Geochemical sampling was carried out in the Key Lake area of northern Saskatchewan in 1973 and 1974. The samples collected were lake waters, organic-rich lake sediments, swamps and some soils. It became apparent that for this particular area the sampling of the organic-rich lake sediments gave the most effective results, both at a reconnaissance level and in detailed work. A very distinct anomalous zone was outlined around the location of the two ore bodies which were



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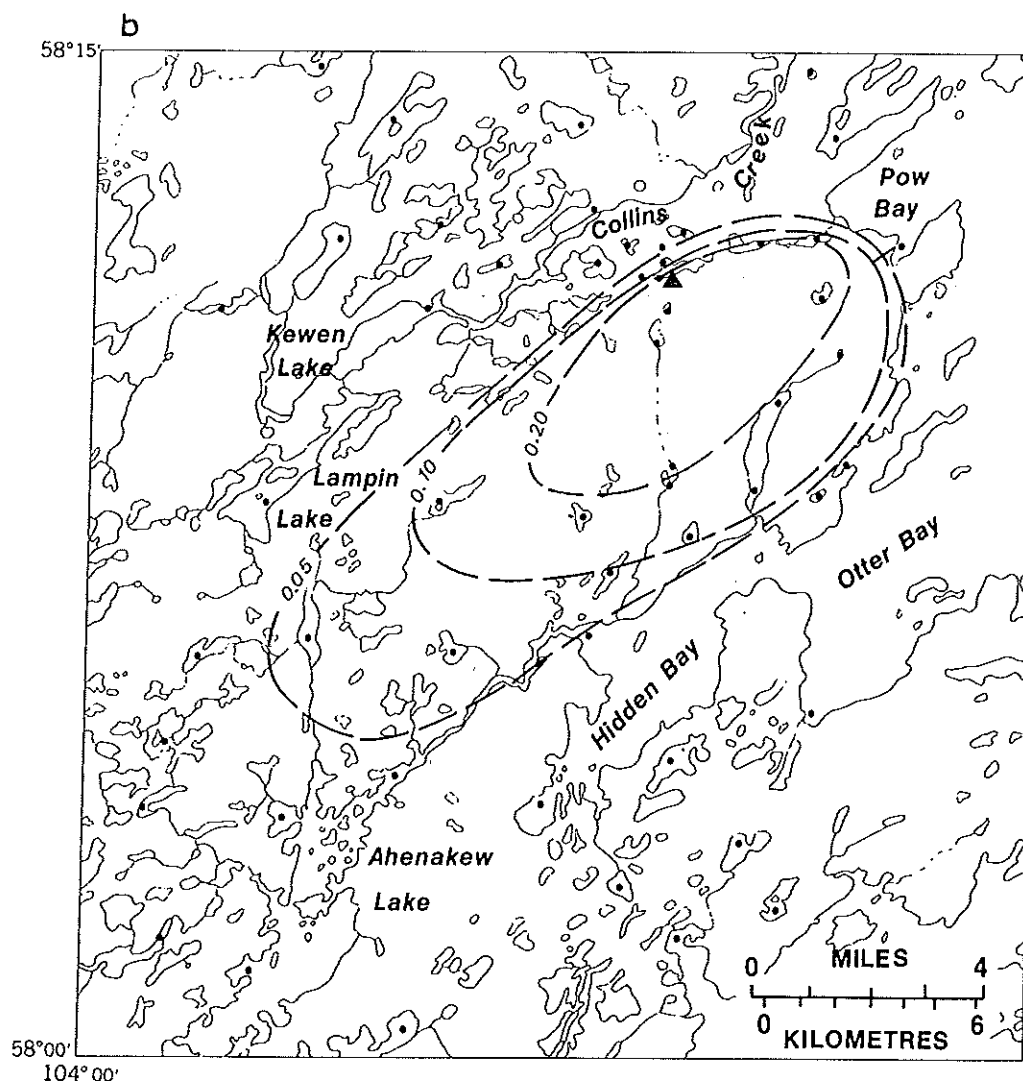


Fig. 13b. Distribution of uranium in surface lake waters as ppb, Rabbit Lake area, Saskatchewan (from Cameron and Ballantyne, 1977).

discovered in the two following years (1975 and 1976)".

The most useful follow-up technique for lake anomalies in glaciated terrain is boulder search and tracing. This may be supplemented by geochemical and mineralogical examination of soils and overburden (Bjorklund, 1976; Bolviken and Gleeson, 1979). For every anomaly that is related to economic mineralization there are many more that are not. In the Canadian Shield the latter are often derived from weakly uraniferous, but extensive, granites and pegmatites. The particular advantage of boulder search is that if mineralized boulders are found, these provide precise knowledge of the nature of the mineralization and the host rock.

Analysis of a more extensive suite of elements is often advocated as a useful follow-up technique. This may involve mobile elements in lake water

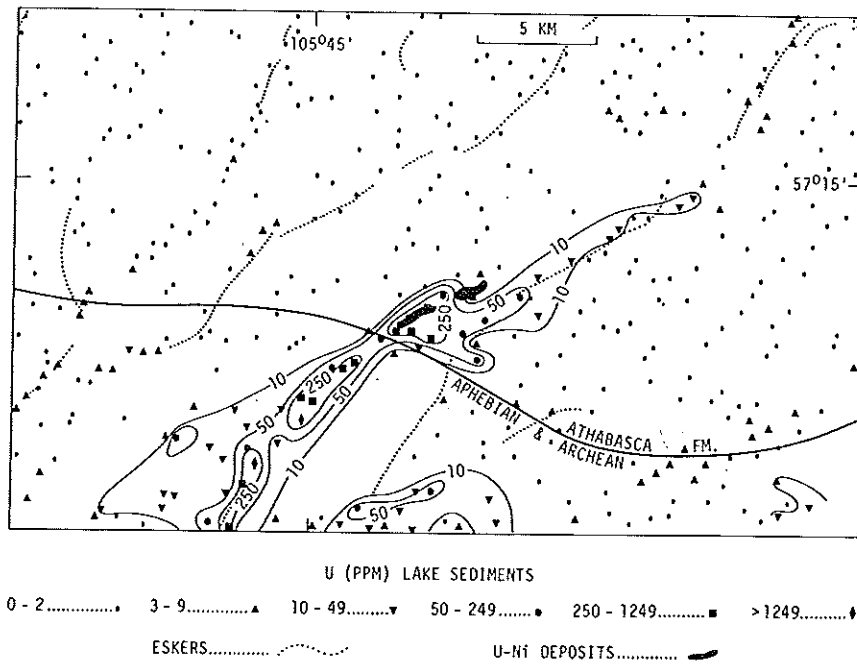


Fig. 14. Distribution of uranium in centre-lake sediments, Key Lake area, Saskatchewan.

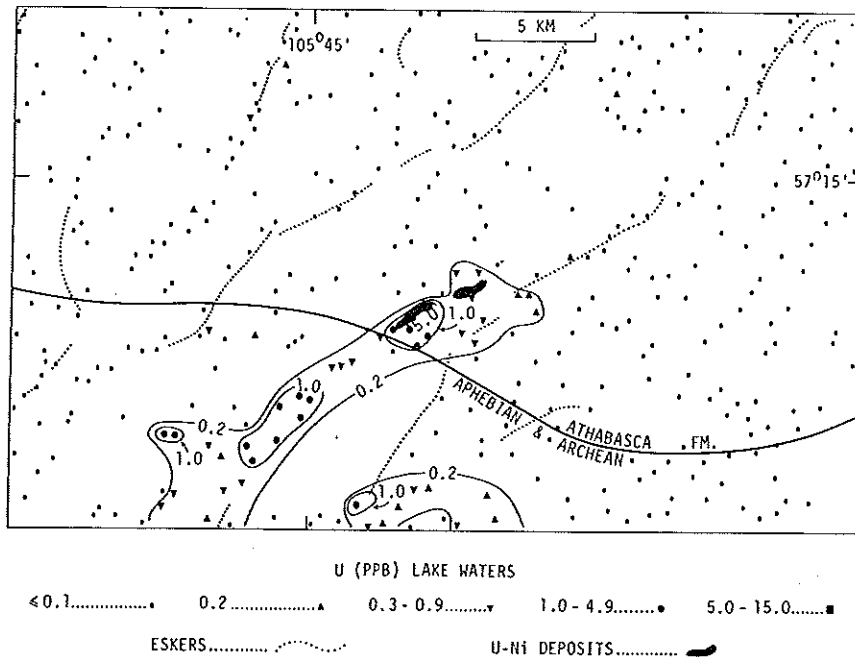


Fig. 15. Distribution of uranium in surface lake waters, Key Lake area, Saskatchewan.

and sediment or less mobile to immobile elements in soil and stream sediment. This approach has had some success in discriminating between anomalies of different origin. For example, Coker (1976) used fluorine in water as a secondary indicator element. However, the usefulness of the approach is limited by the variety of element associations found in uranium

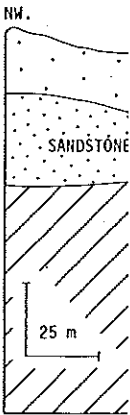


Fig. 16. Gatzweiler

deposits (1976). In this region weakly elevated to the same elevation.

The Key Lake area. Within the area, there are >1000 ppm Sandstone anomalies mineralized.

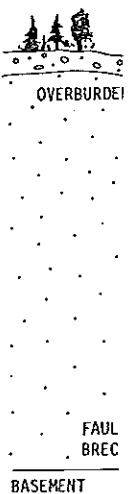


Fig. 17. I scale.

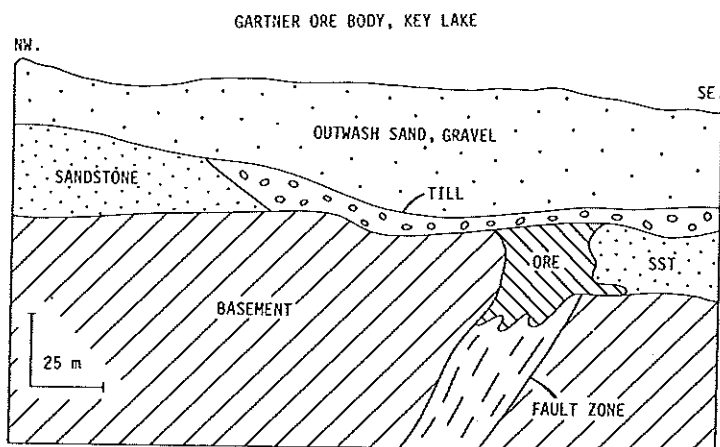


Fig. 16. Geological section through Gartner ore body, Key Lake deposit. (After Gatzweiler et al., 1979).

deposits within a given region (Ruzicka, 1971; Cameron and Hornbrook, 1976). It is imprudent to assume that the undiscovered deposits of a given region will have an element assemblage similar to the known deposits. Also, weakly uraniferous bedrock such as granite or shale, may be enriched in the same elements as are present in uranium deposits.

The Key Lake and Rabbit Lake anomalies are very strong and extensive. Within the former anomaly several sediment samples were obtained with >1000 ppm U, compared to a regional background over the Athabasca Sandstone of <2 ppm. These deposits, with their robust geochemical anomalies, occur near the margin of the Athabasca Basin, where the mineralized unconformity reaches the surface. More recently, other

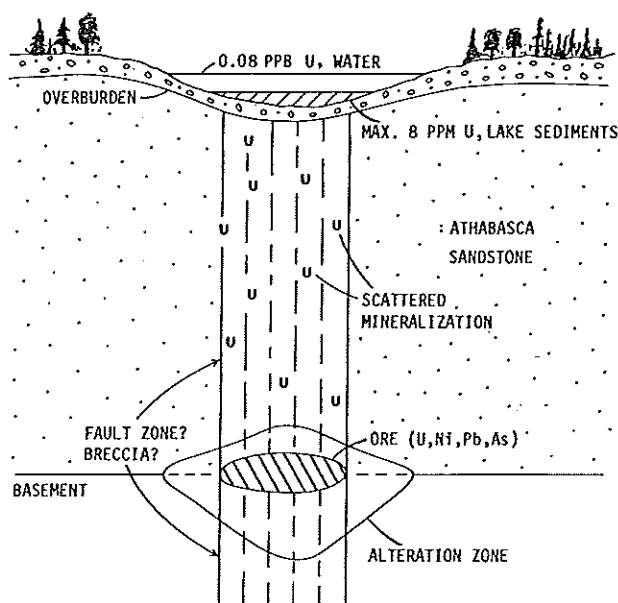


Fig. 17. Idealized section through Midwest Lake uranium deposit, Saskatchewan. Not to scale.

mineralization has been discovered within the basin. A notable example is the Midwest Lake deposit that occurs at a depth of ~ 200 m (Fig. 17). The only mineralization that appears to reach the surface is along a narrow fault zone. While this deposit is of comparable grade and size to Key Lake (2.2 M tons of 1.1% U versus 2.0 M tons of 2.1% U for Key Lake) the depth of burial ensures that the lake sediment and water anomaly is much weaker and less extensive. It appears to be confined to one portion of Midwest Lake, with maximum values of 8 ppm U (sediment) and 0.08 ppb U (lake water) (C.E. Dunn, pers. Commun., 1979). While these values are low in an absolute sense the sediment value is a clear indicator of possible mineralization, since it is four times the regional background. Similarly, uranium in surface water is substantially higher than background. Preliminary data from the area indicates a background of ~ 0.01 ppb U or lower (W.B. Coker, W. Dyck, E.H.W. Hornbrook, pers. commun., 1980).

Gases in lake sediments and waters

For uranium exploration, interest has been directed at two gases: radon and helium. ^{222}Rn is part of the ^{238}U decay series, with a half life of 3.8 days. This limits the distance that it may travel from its parent nuclide, ^{226}Ra , so that detection of a deeply buried deposit will require migration of the parent in groundwater to a near-surface location (Dyck, 1979). Helium in the atmosphere comprises two principal isotopes in the ratio: $^3\text{He}/^4\text{He} = 1.4 \times 10^{-6}$ (Mamyrin et al., 1970). Atmospheric helium is a mixture of "mantle" helium with $^3\text{He}/^4\text{He} \sim 2 \times 10^{-5}$ and "radiogenic" helium with a ratio $\sim 10^{-8}$ (Tolstikhin, 1975). Thus exploration applications may measure neither the total helium content or flux or, better, relative amounts of ^4He (Clarke and Kugler, 1973).

A difficulty inherent in using the gas content of lake waters for exploration is the rapid equilibration between surface waters and the atmosphere.

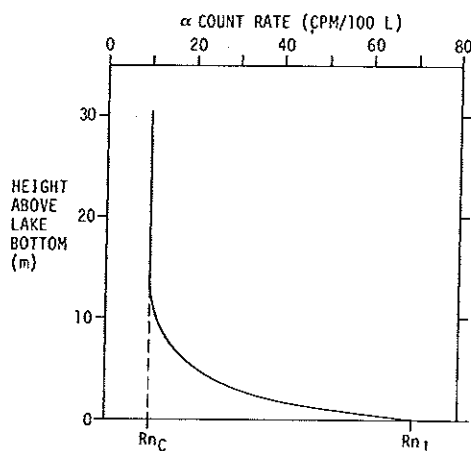


Fig. 18. Vertical profile of radon 222 in Lake Constance, (Modified from Weiss et al., 1978).

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Gases passing through the sediment/water interface decrease in concentration up through the water column as a result of mixing with surface waters, diffusion and also radioactive decay in the case of radon. The rapid decrease in the radon content of the water column is shown for Lake Constance in Fig. 18. The shape of this curve is consistent with mixing by eddy diffusion (Weiss et al., 1978). The content decreases to a limiting value Rn_c , which is the concentration in radioactive equilibrium with radium present in the lake water. In exploration terms Rn_c will be principally determined by the amount of radium entering the lake from its drainage basin, while Rn_I (Fig. 18) will represent the flux from the lake sediments and upwelling groundwaters.

It is apparent from Fig. 18 that slight variation in the distance that bottom waters are sampled above the bottom will cause considerable change in the measured gas content. The mixing of bottom waters with surface waters and the associated decrease in gas content is dependent on a number of factors that are quite variable from lake to lake and with time. These include water depth, wind speed and stratification. For these reasons alternative sampling strategies have been used. These include placing alpha track detectors on the lake bottom (Hogg and Fisher, 1977) or measuring the gas content of the interstitial water of sediment (Dyck, pers. commun., 1979). Another approach is to correct for the effects of varying degrees of mixing of the water column by determining the residence time of the water sample. This is based on the radiotracer pair $^3H \rightarrow ^3He$ (Clarke et al., 1977; Torgerson and Clarke, 1978).

Assuming that it is possible to reliably measure the upward flux of radon and helium into a lake, a further difficulty remains in interpreting the data. The faults along which lakes are often located serve as channels for the ascent of deep groundwaters and gases. These may produce anomalies that are similar to that produced by uranium mineralization.

Clarke et al. (1977) have used measurements of the abundance of 3He , 4He and 3H in Labrador lake waters to locate uranium mineralization. Dyck and Tan (1978) and Dyck (1978) have investigated the helium and radon content of lake waters in the Key Lake area.

CONCLUSIONS

Table II is a summary comparison of the advantages and disadvantages of the three principal materials that may be sampled in northern lakes for uranium exploration. Low-organic, nearshore sediment is the least suitable sampling medium. Of the two remaining media, the principal disadvantage of using waters lies in the analytical methods. At low concentrations of uranium these methods are less precise and more technically demanding than are the best methods of analysing an equivalent abundance of uranium in sediments. But provided that the background level of uranium in the waters of a region is no less than the analytical detection limit, and provided that

TABLE II

Comparison of suitability for uranium exploration of principal lake sampling media

	Low-organic sediments (nearshore)	High-organic sediments (centre-lake)	Surface lake waters
Material provides measure of hydromorphically dispersed uranium	only in part	yes	yes
Within-site variability	high	moderate	low
Seasonal variation	nil?	nil?	moderate
Uranium content influenced by alkalinity	moderate?	little	strong
Sampling cost	moderate—high	moderate	low
Sample preparation costs	moderate—high	moderate	low
Analytical method for	excellent (D.N.C.)	excellent (D.N.C.)	poor precision at low concentration
Suitability for base metal exploration	moderate	good	poor—good

adequate analytical skills and quality control are available, then waters have several advantages. These include low sampling costs, excellent homogeneity within a lake, and minimal requirement for sample preparation. In those parts of the Canadian Shield, and similar terrain, where carbonate rocks are present, it will be necessary to take the alkalinity of the water into consideration when interpreting the uranium in water data.

Centre-lake sediments are moderately more expensive to collect and prepare than waters, particularly if the latter are sampled using special, helicopter-mounted equipment (Cameron, 1978). However, the sediments may be analysed using a thoroughly reliable and precise analytical method, such as delayed neutron counting. Also, over the limited range of alkalinity found in northern humid terrain, no correction is required of the uranium in sediment data for varying alkalinity. Perhaps the principal advantage of sampling centre-lake sediments is that the method is suitable for detecting a variety of mineralization types other than uranium. This particularly applies to organic-rich terrain where base metals seem to be significantly less mobile than uranium.

It appears that organic compounds have a very significant role in the mobilization and dispersion of uranium, as well as in fixing this element. Despite the considerable amounts of money spent on sampling and analysing waters and sediments in the search for uranium, little is known about these processes.

Sampling of northern lakes has very largely focussed on the detection of mineralization within the drainage basin of the lake. In the future increasing

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attention will be given to the search for mineralization beneath the lake. The measurement of gases, such as helium, will be important in this search. However, the development of appropriate methods for detecting hidden mineralization will require much greater financial and technical resources, since orientation surveys must be in three dimensions.

ACKNOWLEDGEMENTS

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