MORSE

URANIUM

Ph.D.



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THE SURFICIAL GEOCHEMISTRY OF RADIUM, RADON AND URANIUM NEAR BANCROFT, ONTARTO WITH APPLICATIONS TO PROSPECTING FOR URANIUM

by

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ABSTRACT

A rapid analytical method for radium-226 (uranium-238 decay series) in sediment and soil, suitable for geochemical prospecting for uranium, has been developed. The method discriminates against potassium and the thorium decay series. Radon emanation into air is less efficient than into water. The following conclusions are based on a study of 90 square miles near Bancroft, Ontario, Canada (Latitude 45°, Longitude 78°).

Radium and uranium in sediments are more useful in prospecting than are radon and uranium in water. Clastic and organic sediments are both useful but clastic are superior. In sediment surveys radium and uranium are equally useful. In reconnaissance prospecting using soils, radium and uranium in A and B horizons are all useful, and in detailed prospecting, all except uranium in the A horizon are useful. Radium is highly preferable to uranium if weathered rocks are used.

Radon in surface water and groundwater has a local source, normally within a few hundred feet. There is not sufficient radium-226 in surface water at Bancroft to account for the radon-222. Radon below the water table does not generally move as a gas. Radium-226 in sediments in the Bancroft area is high enough, on the average, to account for all the radon-222 in water. Results of stepwise multiple linear regression support a model in which radon-222 is added to surface water by decay of radium-226 in stream sediments and by influx of groundwater, and lost by aeration and radioactive decay. Organic material reduces the radon-emanating efficiency of sediment.

Uranium and, to a lesser extent, radium, are concentrated by organic sediments. Radium is higher in the A horizon of soil than in

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the B; uranium is not. Uranium is depleted relative to radium-226 in clastic sediments and weathered rocks. Radium and uranium in sediment and radon in water have lognormal distributions.

ACKNOWLEDGEMENTS

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CHAPTER 1 INTRODUCTION

STATEMENT OF THE PROBLEM

The problem comprises two aspects. The first is to test the applicability of the geochemistry of radium-226 (from the uranium-238 decay series) to prospecting for uranium. To do this it was required to (1) develop a suitable analytical method, (2) carry out a geochemical survey by collecting geochemical samples (water, sediment and soil), analyzing them for radium-226, radon-222 and uranium, and comparing the distribution of radium-226 with that of radon-222 and uranium as related to known uranium deposits and bedrock geology. The second aspect is to determine so far as possible the history of the radon-222 observed in water, that is, to determine where the transmutation of radium-226 to radon-222 takes place, and hence whether radon-222 arrives at its present location as the element or in the form of one of its precursors.

Radium-226 can definitely be applied to prospecting for uranium although the advantage of this method is slight relative to previously available methods using radon and uranium. A model is proposed in which the level of radon in surface water is controlled by addition of radon through decay of radium-226 in the sediments and influx of radon-charged ground-water, and by loss of radon due to aeration and radioactive decay. Results of stepwise multiple linear regression analysis of data collected for 182 stream sample points are consistent with, and hence lend support to, this model.

Work began in the summer of 1968 as part of a Geological Survey

of Canada project 670030 led by A. Y. Smith (Smith and Dyck, 1969).

Sediment samples collected by the writer in 1968 were analyzed for radium-226 in the fall of 1968 at Queen's. Results of this early work showed that measurement of radium-226 in sediments provides a reconnaissance tool for uranium prospecting. This information was considered significant enough to warrant immediate publication (Morse, 1969, a and b).

Work continued in the summer of 1969 as project 690080 led by the author. A comparison of radium-226, radon-222 and uranium as geochemical indicators for uranium (Morse, 1970) was largely summarized from this thesis.

THE URANIUM INDUSTRY

Uranium cres were first used as a source of radium for medical purposes. During the 1940's and 1950's, exploration and production source in response to the military demand. In the 1960's, as the military demand declined, free world production dropped from a high of 44,000 tons of 0.08 in 1959 to 20,000 tons in 1966.

The electric generating industry will soon require large amounts of uranium. The most recent forecast of uranium requirements known to this writer was made in January 1969 by the European Nuclear Energy Agency and cited by Williams (1969). This study predicts that the annual non-communist demand will rise from 12,500 tons U_3O_8 in 1968 to between 73,000 and 106,000 tons in 1980. Williams (1969) has made an exhaustive study of Canada's future in uranium supply. On the basis of reserves and production capacity, and the above prediction, he concludes that new discoveries must be made and brought into production by 1973 or 1974, if

the price is to be kept below \$10 per pound U_3O_8 .

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In view of the lag time between discovery and production, it is essential that intense exploration be carried on now. This is recognized by both the mining and petroleum industries, and the non-communist world is now experiencing an exploration rush. Any improvement in exploration technology will result eventually in lower prices or a more secure supply of uranium. Such an improvement is the purpose of this thesis.

GEOCHEMICAL PROSPECTING FOR URANIUM

A number of elements have potential application to prospecting for uranium. They comprise members of two uranium decay series as well as products of natural fission of uranium. In this thesis, consideration is restricted to the nuclides radium-226 and radon-222, and the element uranium. Geochemical prospecting methods using these materials are not affected by radiation from the thorium decay series or from potassium (see Chapter 4) as are some geophysical methods. Uranium and radon-222 have received much attention in the past, and helium has been used (Hawkes and Webb, 1962, pp. 366, 371 and 375). Radium-226 as a geochemical prospecting tool for uranium has largely been neglected, and is a main subject of this thesis. Application of the other members is suggested as a subject for further research.

Development of a rapid analytical method for radium-226, from the uranium-238 decay series, is an important contribution of this thesis. Even though the applicability of radium geochemistry to prospecting for uranium was publicized in May, 1969, (Morse, 1969a) it is still of no use to mining companies because the commercial laboratories cannot provide inexpensive analytical services.

As a sampling medium, any natural material can potentially be used. This thesis however is limited to water, sediment and soil. J. L. Walker (1968) warms that organic material in sediments should be avoided because of interference with analytical methods and highly erratic metal contents. At many places in the Bancroft area, the sediment is totally organic, and clastic material is not available. Walker's reservation is tested in this thesis in the case of uranium and radium, and a statistical technique of mixing the results obtained from organic sediment with those obtained from clastic sediment is proposed. Walker (1968) goes on to note that "...water sampling and analyses will work by exception rather than rule, and even in these circumstances this technique is generally a cumbersome method of stream sediment sampling." Results of this thesis show the hydrogeochemical methods are indicative, but not as effective as stream sediments.

PHYSICS OF THE URANIUM-238 DECAY SERIES

Most of the naturally occurring radioactive nuclides are members of one of three radioactive decay series (see Tables 1,2 and 3). The parents of these series are uranium-238, uranium-235 and thorium-232. The parent nuclide decays into a radioactive daughter nuclide, which in turn decays, and so on, until a stable nuclide (lead) is formed. In addition to the elements shown in Tables 1, 2 and 3, several of the transformations involve emission of an alpha particle or helium nucleus. The

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TABLE 1

Uranium-238 Decay Series

Principal members only; isotopes constituting less than 0.2 per cent of the decay products are omitted.

Uranium-238 (4.51 \times 10 9 y) Thorium-234 (24.10d) Protactinium-234 (1.14m) **↓** β Uranium-234 (2,48 \times 10⁵y) 4 α [Thorium-230 $(8.0 \times 10^{4}y)$ + α Radium-226 (1,622y) Radon-222 (3.825d) **ψ** α Polonium-218 (3.05m) ∳ α Lead-214 (26.8m) ¥β Bismuth-214 (19.7m) Polonium-214 (1.50 \times 10⁻⁴s) Ψ α Lead-210 (22y) ¥β Bismuth-210 (5,02d) ¥ B Polonium-210 (138d) Lead-206 (stable)

Taken from Lang, Griffith and Steacy (1962).

TABLE 2

Uranium-235 Decay Series

The series of the series of the series

```
Uranium-235 (actino-uranium) (7.13 \times 10^8 y)
                                                                                          ψ α
                                                                                          Thorium-231 (25.64h)
                                                                                          ∳ β
                                                                   Protactinium-231 (3.43 × 10^{4}y)
                                                                                        + a
                                                                                       Actinium-227 (21.7y)
                           (98,8%)β <sup>≠</sup>
                                                                                                                                                                                                \alpha(1.2\%)
Thorium-227 (18,6d)
                                                                                                                                                                                   Francium-223 (21m)
                                                                                        * Constitution Confirms
                                                                                              Radium-223 (11.2d)
                                                                                               oldsymbol{\psi}_{i} , oldsymbol{lpha}_{i} , oldsymbol{lpha}_{i}
                                                                                                Radon-219 (3.92s)
                                                                                             \Psi_{0}(oldsymbol{lpha})
                                                                            Polonium-215 (1.83 \times 10<sup>-3</sup>s)
                                                                                                   * a
                                                                                                   Lead-211 (36.lm)
                                                                                                January Harbert
                                                                                              Bismuth-211 (2.16m)
                         (99.68%)a 4
                                                                                                                                                                                 _{\lambda} _{\beta}(0.32\%)
Thallium 207 (4.76m)
                                                                                                                                                                                   Polonium-211 (5.2 × 10^{-1}s)
                                                                                                   The section of the se
                                                                                                  Lead-207 (stable)
```

Taken from Lang, Griffith and Steacy (1962).

TABLE 3

Thorium Decay Series

```
Thorium-232 (1.39 \times 10<sup>10</sup>y)
                                                                                                                                             Radium-228 (6.7y)
 Two price of the many of the second of the s
                                                                                               Actinium-228 (6.13h)
                                                                                                                                         Thorium-228 (1.90y)
                                                                                                                                       Radium-224 (3.64d)
                                                                        Radon-220 (54.5s) (popularly called thoron)
                                                                                                                                  Polonium-216 (0.158s)
                                                                                                                                             Lead-212 (10.6h)
                                                                                                                                   ß
                                                                                                                                       Bismuth_212 (60.5m)
                                                                                                                                                                                                                                                                                \alpha(33.7\%)
                                                     (66.3\%)\beta
Polonium-212 (3.0 × 10^{-7}s)
                                                                                                                                                                                                                                           Thallium-208 (3.1m)
 all and the second of the second
                                                                                                                               Lead-208 (stable)
```

Taken from Lang, Griffith and Steacy (1962).

latter soon picks up two electrons to become atomic helium. Natural uranium is 99.27% U^{238} , 0.720% U^{235} , and 0.006% U^{234} . Natural thorium is 100% Th^{232} .

If the parent of the series is separated from its daughters and then the series is left undisturbed, then, after a time, the whole series reaches equilibrium; or rather, approaches it asymptotically. In other words, each member of the series decays at the same rate. Equilibrium is attained in the $\rm U^{238}$ series in about 1 million years; in the $\rm U^{235}$ series in about 100,000 years; and in the thorium series, in about 100 years. All three series are in equilibrium in most ores and rocks; however, in young or recently weathered rocks, and in soils and sediments, the $\rm U^{238}$ series especially can be out of equilibrium.

The amount of a particular nuclide present at any given time after the parent is separated from the rest of the series can be found by solving the equations derived by Bateman (Kaplan, 1962). Solutions of these equations for the U²³⁸ series down to radon for several time periods are presented in Table 4. The units used, equilibrium units, are the number of atoms times the appropriate decay constant, assuming one equilibrium unit of U²³⁸ initially. Equilibrium units, in other words, express the amount of nuclide present as a proportion of that present at equilibrium. Six of the entries are expressed as maxima as the Bateman equations cannot give an answer less than 0.01 equilibrium units. Maxima less than this are found by assuming that U²³⁴ was at its final level for the whole period and solving the Bateman equations for the shorter series.

It is clear from Table 4 that any significant correlation between U²³⁸ and its daughters below Pa²³⁴ in post-Pleistocene material

TABLE 4

Amount of Nuclide Present Starting with Pure U²³⁸ -- Equilibrium Units

	Initial		10,000 yrs.	100,000 yrs.	1,000,000 years
_238	1	1	1	1	1
Th ²³⁴	0	1	1	1	1
Pa ²³⁴	0	1	1	1	1
_U 234	0	.003	.026	.24	.938
Th ²³⁰	0	< 3 × 10 ⁻⁵	< .002	.08	.907
Ra 226	0	<10 ⁻⁵	< .0016	.08	.907
Rn ²²²	0	< 10 ⁻⁵	< .0016	.08	.907

indicates similar geochemical behavior, not radioactive production. For example, radon in fresh water arrived there by some means other than the decay of uranium in solution.

Another approach to the physics of the uranium decay series is to consider the degree of equilibrium attained by parent and daughter pairs. Equilibrium obtains when the parent is much longer-lived than the daughter (see Appendix I). The degree of equilibrium is expressed as a ratio

$$\frac{N_2\lambda_2}{N_1\lambda_1} = 1 - e^{-\lambda_2 t}$$
 (Equation 3, Appendix I),

or, if multiplied by 100, as a percent. Radon-222 is 50% in equilibrium with radium-226 in 3.8 days, 90% in 13 days and 99% in 25 days. Radium is 50% in equilibrium with thorium-230 in 1600 years, 90% in 5500 years and 99% in 11,000 years. The rapid approach to equilibrium of the pair radium-radon is the basis of the analytical method for radium discussed in Chapter 3. The same fact indicates that in nature radon-222 will not be far removed in time from its parent radium-226.

APPLIED GEOCHEMISTRY OF URANIUM

Work on the applied geochemistry of uranium up to 1960 has been summarized by Hawkes and Webb (1962). In their words, uranium is "extremely mobile under alkaline, oxidizing conditions...The uranium content of fresh water is extremely effective as a method of reconnaissance exploration..." The same authors note the affinity of uranium for or-

ganic matter in peat bogs.

These principles have since been tested in the Bancroft area by Chamberlain (1964) who measured uranium in over 1100 water samples and a few sediment samples over an area of 1850 square miles. He found the greatest potential use of the method to be in reconnaissance prospecting. Of detailed prospecting he notes:

On a local scale, anomalously high uranium values in waters associated with orebodies generally decrease to near-background level within a few hundred feet. Results indicate that uranium is extracted from solution by the reducing effects of decaying organic rationals. In the investigated area, and presumably over much of the Canadian Shield, uranium hydrogeochemistry thus offers only restricted guides to specific ore targets.

The usefulness of uranium in surface waters as a reconnaissance prospecting tool has been further substantiated by Smith and Dyck (1969) at Bancroft, Ontario, MacDonald (1969) at Beaverlodge, Saskatchewan and Meyer (1969) in Labrador. Either lakes or streams can be used. Lakes are particularly useful in the Canadian Shield due to their ubiquity and the simplicity of sampling them by aircraft. Reconnaissance samples can be collected at the rate of ten or more per hour. Meyer (1969) reported a 3- to 4- fold increase in uranium content of lakes from July to September, but MacDonald (1969) found no such variation.

Uranium in residual soil has been applied extensively to uranium prospecting (Hawkes and Webb, 1962). "Analysis of plants for uranium has been one of the most successful of the geochemical methods used on the Colorado Plateau."

APPLIED GEOCHEMISTRY OF RADON

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With an atomic number of 86, radon is the heaviest noble gas.

Radon-222, the most abundant and longest-lived isotope, has a half-life

of 3.8 days,

The solubility coefficient of radon in water at various temperatures, taken from Sedlet (1966), is given in Table 5. At 20°C, under equilibrium conditions, the concentration of radon in water will be 0.25 times the concentration in the gas phase. The abundance of radon in the earth is extremely low and below the water table, in the absence of a gas phase radon must exist in either the dissolved or the adsorbed state. Unless a mobile gas phase is present—and this is extremely unlikely in Precambrian terranes—the mobility of radon below the water table is limited by the mobility of the groundwater in which it is dissolved. Because groundwater moves under normal conditions at the rate of only a few feet per day, radon will not normally move more than a few hundred feet before it has decayed to an undetectable level. The high levels of radon—222 frequently reported in spring and some mine waters are due to radium—226 in the solid material of the aquifer, either by itself or in uranium minerals.

It has been suggested several times to the author that uranium deposits below lake bottoms in the Canadian Shield might be detected by measuring the radon dissolved in the water at the bottom of the lake. It is clear from the foregoing that radon will not "percolate" upwards from the uranium deposit as a gas phase, but will move in solution with the groundwater.

In surface water, nonequilibrium conditions prevail. Movement of air above the water will ensure removal of radon in the gas phase so that here its concentration is negligible compared with that for equilibrium. Therefore, concentration of radon in surface water will be dependent upon kinetic factors such as diffusion and turbulence which contribute to the movement of radon to the water—air interface. The effect of turbulence on the level of radon in surface water is considered

in Chapter 7. Hawkes and Webb (1962) state that radon is "extremely mobile as a dissolved constituent of water..." As shown above this cannot be true because (a) below the water table the short half-life together with the slow groundwater movement precludes its movement over great distances, and (b) escape into the air will be rapid. (In a study of radon in small streams in the Wasatch mountains, Rogers (1958) observed that radon levels dropped off to zero within as little as 500 feet downstream from the point of influx).

In the Bancroft area raden-222 is found in high concentrations several miles downstream from its ultimate source, uranium minerals. Its dispersion is greatly increased by the dispersion of its parent radium-226. The source of radon in surface water is one of the problems considered in this thesis.

The usefulness of radon in surface water as a reconnaissance prospecting tool for uranium deposits has been demonstrated by Smith and Dyck (1969) and Dyck and Smith (1968). As in the case of uranium, either lakes or streams can be used with the same advantage of rapidity of sampling.

Above the water table radon is more mobile because in this case it is in the gas phase. The applied geochemistry of radon in soil has attracted considerable attention, most recently by Dyck (1969b). He has

TABLE 5*

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Solubility of Radon in Water

在整体的,1000年的中央中央管理的1000mm,1000年

Temperature	Solubility Coefficient**
	the Marina Marinet Const.
18° C	,2 9
des 4, 20°, C	.25
: 	.17

* After Sedlet (1966)

Allegated Management of the contract of the con-

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Transfer on Are

** The solubility coefficient is the ratio of the concentration in water, in mass per volume to that in the gas phase, in mass per volume, at a particular temperature.

reported that radon determinations in soil have outlined uranium-bearing rocks in the Gatineau Hills, Quebec, and the Elliot Lake, Ontario areas (Dyck, 1969b), and the Bancroft, Ontario area (Dyck, 1968).

APPLIED GEOCHEMISTRY OF RADIUM

The geochemistry of radium in natural waters is probably controlled by coprecipitation with barium sulfate, by adsorption to sediment or by binding with organic matter rather than by solubility of radium itself. Starik (1963) has shown that radium in the ocean is adsorbed to sediments. This is in addition to the well-known concentration of radium in ocean sediments due to the precipitation of its insoluble parent thorium-230. The affinity of radium for organic matter in peat has been known to the Russians since 1943 (Titayeva, 1967). It is confirmed in this thesis (Chapter 5).

The applied geochemistry of radium has largely been neglected. Hawkes and Webb (1962, p. 371) mention that "radium deserves investigation as a pathfinder for uranium." Titayeva, 1967, Vinogradov, 1959, and other Russion scientists have studied radium in soil and sediment. One would suppose that they have tested its application to prospecting, but this writer was unable to find any specific mention of it in the available literature. Grimbert (personal communication) at the Commissariat a L'Energie Atomique in France has studied the geochemistry of radium in soils in an effort to apply it to prospecting, but has not published any results, and it is understood has no immediate plans to do so.

Chew (1956) approached the subject of the applied geochemistry of

radium when he measured, with a Geiger counter, radioactivity in modern stream gravels in the Colorado Plateau and found high values downstream from mines. Because most of the radioactivity of uranium minerals is due to radium-226 and its daughters, and because the uranium deposits in the Colorado Plateau are poor in thorium, it is apparent that the increase in radioactivity associated with the uranium deposits was due to radium. Background levels were, no doubt, in part due to thorium and potassium minerals.

In Canada it is frequently desirable to discriminate between radioactivity due to uranium and that due to thorium and potassium. Thorium deposits are common in both the Bancroft and Elliot Lake camps and cause radicactive anomalies that cannot be distinguished from those due to uranium. The ubiquity and irregularity of glacial materials rich in potassium and thorium add to the confusion. Chew's method would be unsatisfactory in this case. Determining radium—226, the method developed in this thesis, is, on the other hand, specific for uranium.

This thesis has two purposes: (1) to evaluate the usefulness of radium geochemistry per se as a tool for uranium prospecting and compare it with other methods, and (2) to determine as much as possible the history of the radon-222 observed in water, that is, to determine where the transmutation of radium-226 to radon-222 takes place, and hence whether radon-222 arrives at its present location as radon-222 or in the form of one of its precursors.

The first goal was approached by collecting drainage samples (water and sediment) over a 90 square mile area; analyzing the sediments for radium and uranium and the waters for radon, uranium and radium; and comparing the response of the different elements in the two media to

known uranium deposits and rock type. In addition, a small collection of soils was analyzed for radium and uranium. This determines which of the methods works and which works best. A suitable analytical technique for radium was developed to accomplish this.

Progress towards the second goal, the source of radon-222 in water, comes about largely as a byproduct of the first. Consideration is given to the emanation of radon-222 by sediments. This is also the basis of the analytical method developed for radium. Then the effect of several variables on the radon content of surface water is examined.

CHAPTER 2 AREA STUDIED

An area of 90 square miles at Bancroft, Ontario was selected for the following reasons. It contains several uranium deposits. Three of these have supported producing mines and their lateral extent is well known. The area comprises two different geological terranes, the Haliburton-Hastings Highlands gneiss complex and part of the Hastings Basin area of marble, paragneiss and amphibolite. A modern geological map on a scale of one inch to one half mile is available.

Mining activity ceased in 1964 and exploration ceased even earlier, reducing the chances of contamination. The drainage downstream from Bicroft and Faraday mines may have been contaminated by ore processing activity, but this drainage can be left off the geochemical maps without destroying the sample coverage. Farms are uncommon and most of the area is nearly pristine.

In contrast to some areas of the Canadian Shield, streams and stream sediments are abundant. In spite of the ubiquity of outcrop, some areas of soil are available for pedogeochemical studies, notably over Bicroft and Fáraday mines.

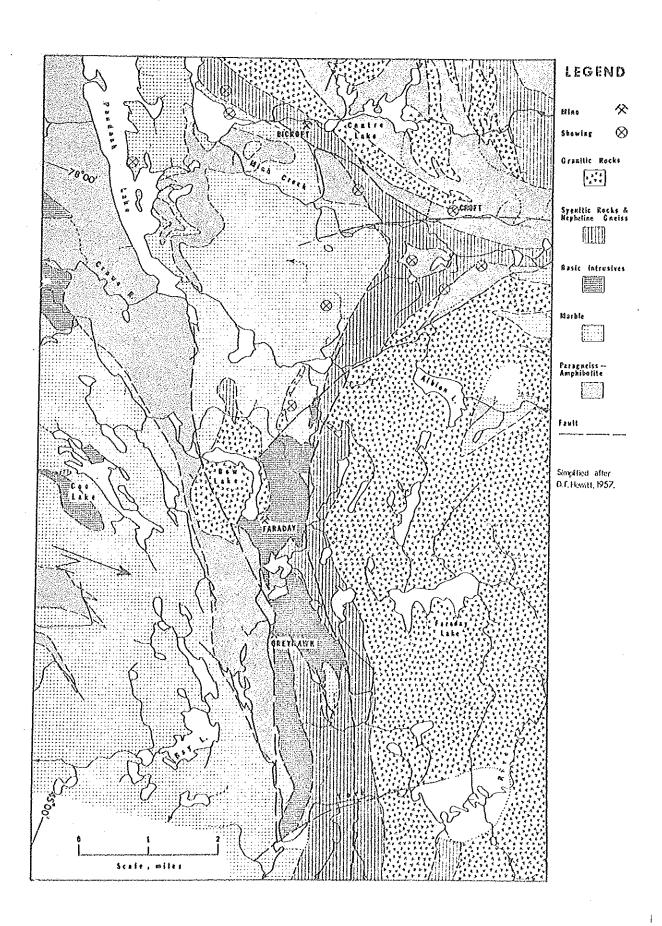
The area is easily accessible from Kingston or Ottawa by a two or three hour drive. Most of the sample points can be reached by automobile or by a short walk. This allowed the author to collect each sample and make observations personally.

LOCATION AND ACCESS

The area studied (Figure 1) comprises a rectangle of 90 square

FIGURE 1

Geology and Uranium Occurrences, Bancroft Area



miles, 12 miles by 7.5 miles. It covers parts of four National Topographic map sheets, 31C/13 west, 31D/16 east, 31E/1 east and 31F/4 west; and parts of Faraday township of Hastings county and Cardiff township of Haliburton county.

Access is provided by five paved highways, a railway and an airport. A dense network of roads covers the area; no point is further
than 1.3 miles from a passable road or trail. York River and several
lakes provide access by boat to some of the more remote locations.

GEOLOGY

The area lies in the Grenville Province of the Canadian Shield.

The Grenville front is 160 miles to the northwest and the Paleozoic cover rocks are 30 miles to the south.

The rocks, all of Precambrian age, are of two main types (see Figure 1): (1) Grenville type metasediments, mainly marble, paragneiss and amphibolite to the south; and (2) plutonic rocks, mainly granite, syenite and gabbro, together with their gneissic and hybrid equivalents to the north. These two areas are separated by a narrow band of syenitic rocks and nepheline gneiss (Hewitt, 1959).

Hewitt (1959) feels that the Grenville metasediments are the oldest rocks in the area. These were then intruded by gabbro and diorite, followed by nepheline syenites and then by syenites and granites.

The structure is dominated by two areas of plutonic rocks, the dome-like Cardiff plutonic complex, part of which is included by the northwest corner of the area, and the south-dipping Faraday granite sheet

which occupies the northern portion of the area. Between these two plutens, the metasediments, syenitic rocks and nepheline gneiss have been plastically deformed into a V-shaped synclinal reentrant. Similar domes and reentrants are common nearby. Numerous steep faults cut the area.

URANIUM MINES AND SHOWINGS

In an area 40 miles square centered 20 miles southwest of Bancroft are no less than 122 uranium occurrences (Ontario Department of Mines, Map No. 1957b). Only 4 of these have produced economic amounts of ore. They lie in a straight line stretching from 3 to 12 miles southwest from Bancroft.

The area studied contains 3 of the 4 abandoned mines and ten of the uranium occurrences. All these deposits are in or near the narrow strip of syenitic rocks and nepheline gneiss which lies between the granitic rocks on the north and the metasediments on the south. That this distribution is true in general can be seen on the Ontario Department of Mines map of the larger Haliburton-Bancroft area (Map No. 1957b). The uranium deposits are of several types, but all the successful mines are in complex bodies of granitic or syenitic pegmatite. The following brief descriptions are summarized from Satterly (1957).

The Bicroft ores are in a north-south zone of granitic bodies occurring in a band of syenitized paragneiss and amphibolite. The zone is 16,000 feet long and extends to the Croft workings (see Figure 1).

At Faraday the radioactive minerals occur in bodies of leucogranite, leucogranite pegmatite, and pyroxene granite (or syenite) pegmatite, cutting metagabbro and gabbroic amphibolite." (Satterly, 1957, p. 110) Ore has been developed in a zone extending 2500 feet southwest from the shaft.

The Greyhawk ores are in pegmatite in a northeast striking body of diorite and metagabbro (see Figure 1). Development was limited to an area extending 500 feet east and northeast from the shaft. No ore has been found south of the fault shown in Figure 1 passing through the mine area.

PHYSIOGRAPHY

Elevation ranges from 1050 to 1700 feet. The area is divided into two physiographic terranes. The north half, underlain by granitic rocks and characterized by high relief, is part of the Haliburton Highlands. The south half, underlain by metasediments with scattered basic intrusives and characterized by lower relief and lower elevation, is part of the Hastings Basin.

Nothing has been published on the Pleistocene geology. The glacial features, as observed by the writer, are similar to those in the rest of the Canadian Shield, namely, polished rock surfaces with little chemical weathering, abundance of glacial till and erratics, dislocation of drainage and numerous deposits of sand and gravel. Ice movement direction in nearby areas was 15 degrees west of south (Geological Survey of Canada, Map 1253A). Ice retreated from the general area about 12,000 years ago (Geological Survey of Canada, Map 1257A).

Two rivers, the Crowe and the York, drain the area. The eastern

two-fifths drains to the east to the York River, the larger of the two. The York flows north to join the Madawaska which then flows into the Cttawa. The remainder of the area is drained to the south by the Crowe River. The Crowe flows south into the Trent which then flows into Lake Ontario.

Swamps and lakes abound; there are over a hundred lakes large enough to show on a 1:50,000 scale map. Individual streams alternate between fast-flowing stretches with rocky beds and slow-moving swampy areas. Beaver dams are abundant. The writer encountered 17 springs; this suggests that they are relatively common.

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SOIL AND VEGETATION

A soil survey of Hastings County, which includes Faraday township, has been published (Gillespie, 1962), but none is available for Cardiff township. However, because three quarters of the area studied lies in Faraday township, the following discussion, condensed from this report, is useful.

Seventy-five percent of the area is mapped as rockland. This is described as being "50 to 90% rock or thinly covered rock with small deposits of deeper soil materials in the crevices." Most of the remainder is mapped as sandy loam and loamy sand. Organic "muck" occupies several small patches, and bottom land (recent alluvial deposits) occupies one small patch. Except for muck, bottom land and a small patch of gleysolic soil, the soils are all podzolic.

The area is covered with forest except for scattered patches of

farmland, swamps and rock outcrops. Areas of mixed hardwoods and conifers are most common. These grade into pure hardwoods on one hand, and pure conifers on the other. The common hardwoods are maple, poplar, birch, elm and oak; the colifers are cedar, spruce, fir, hemlock and pine. Many abandoned farms have been planted to pine.

CLIMATE

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The Canada Department of Transport (D.O.T.) maintains a weather station at the village of Bancroft where maximum and minimum temperatures and precipitation are recorded daily. Monthly and yearly average of these measurements for part of the period 1931 to 1960 have been published (Canada Department of Transport, 1967). Comparative data for the period of this study were provided by the Ottawa and Toronto offices of the D.O.T. These data are presented in Table 6, Snow accounts for about 20% of the total yearly precipitation. Table 6 shows that 1968 was a normal year, but 1969 was wetter and colder than usual. Precipitation in the first seven months of 1969 was 20.50 inches compared with 17.09 for the normal year.

CULTURE AND CONTAMINATION

The area was first settled in the mid-nineteenth century. Many of the farms were abandoned within a few years, but a few scattered dairy farms remain. Logging continues to be an important industry; the original pine forests were logged off early and gave rise to second

TABLE 6

Temperature and Precipitation at Bancroft

	Mean Tem	erature (d	degrees F)	Precipitat	tion (inche	es of water)
Month	Normal	1968	1969	Normal	1968	1969
Jan	14,1	•		2.59	1.59	1.47
Feb	14.1		en e	1.68	2.19	.60
Mar	23.7			2.63	2.46	1.16
Apr	37.3	·		2.31	1.16	3.93
May	51.7			2.42	1.69	4.51
June	60.9	60.8	59.3	2.61	4.44	6.07
July	65.3	65.6	64.5	2.85	1.82	2.76
Aug	63.5	62.3		2.41	2.20	·
Sept	54.4			3.31	4.68	
Oct	43.0			2.64	2,25	
Nov	31.4			2.93	3.53	
Dec	18.1			2.76	3.90	
Year	39.8			31.14	31.91	

growth hardwoods. In recent years tourism has become perhaps the most important industry. Tourists are attracted by opportunities for fishing, hunting, boating, and mineral collecting. Mining activity boomed during the nineteen-fifties but ceased altogether in 1964 when the uranium demand fell. The Faraday mine, the last to close, has a large amount of ore and is in a position to resume production given a favorable sales contract.

Ore processing at the Bicroft and Faraday mines may have added uranium and its daughter products to Paudash and Bow Lakes, respectively, and to the streams draining them. Ore was not processed at Greyhawk mine, although waste dumps and mine workings may have affected natural distribution of these elements.

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CHAPTER 3 SAMPLING

DRAINAGE SURVEY

Drainage samples were collected at 258 points over an area of 90 square miles (Chapter 2). Sample sites were selected, in general, on the basis of accessibility and uniform density; however, some detailed sampling was carried out around Bicroft and Greyhawk mines. If the latter samples are omitted along with samples from Paudash and Bow Lakes and the streams draining them, which may be contaminated (see Chapter 2), a "random" sample of 221 points results.

Water and sediment samples were collected from all types of surface water bodies: streams, lakes, swamps, ponds and springs. Stream samples were collected as near the center as possible. Lakes, swamps and ponds were sampled near the edge except for two samples in Faraday lake, one in Coe Lake and eleven in Faudash Lake which were collected in a boat. Sediment at these locations was collected by dredging with a bucket on a rope. The maximum depth sampled was 37 feet.

Water samples were collected in eight-ounce (230 cc) glass bottles by partially submerging the bottle and allowing it to fill with a mini-mum of turbulence. Sediment samples were collected by hand and placed in Kraft paper envelopes.

Two types of sediment were found: clastic sediments which consist of sand, gravel and silt; and organic sediments which consist of dark-colored, soft, unconsolidated organic material. Where present at the same location, the two types were always separate and distinct.

Both types were collected at every sample point at which they were found together. About a third of the sample locations had both types of sediment, a third had only organic and a third had only clastic material. At 37 locations bottom material consisted of a mixture of organic sediment and plant parts, both living and in various stages of decay. This material was sampled and treated as sediment, but the results are treated separately in Chapter 5 and Table 17.

For each sample, an 80-space geochemical sample information card was filled out in the field. Of the information recorded, all of which is listed in Appendix V, the following observations are relevant to this thesis: sample type, width of stream, depth, rate, turbulence, composition of sediment, possibility of contamination, water temperature and pH. Except for pH, which was measured with a Beckman Model N pH meter, and temperature, all the observations were visual estimates.

SOIL SURVEY

Eighty-two soil samples were collected from four areas. Two lines were run over Faraday mine and one over Bicroft mine. Six samples were collected from a small area well away from known mineralization to give an indication of "background." The Bicroft line is oriented eastwest with its zero point 720 feet north and 300 feet east of No. 1 shaft. The Faraday east line (Figure 21) has its origin on a road at 15,109 feet north and 14,405 feet east (mine grid) and is oriented at N 33° W. The Faraday west line (Figure 22) runs along the 13,000 foot east line from 14,250 to 14,850 feet north (mine grid). The writer is indebted to

A. Y. Smith who established the picket lines.

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Two soil horizons were found. The A horizon, normally one to two inches thick, is black and rich in organic material. The underlying B horizon is rusty brown. No leached zone is visible in the A horizon. In swampy areas the rusty brown layer is not present, and black soil extends to at least two feet. In most areas soils are thin and stony. Outcrop is abundant. Maple forests cover the area of the soil surveys.

Samples were collected at fifty foot intervals wherever the soil was at least a foot deep. Both A and B horizon were sampled, A from just below the litter and B from a depth of about a foot. Where the rusty B horizon was not present, only a sample of the A horizon was taken.

CHAPTER 4 ANALYTICAL METHODS

Bancroft the following day or at least within a few days of collection.

Nearly all the water samples were analyzed for uranium. Those collected in 1968 were analyzed in a field laboratory at Elliot Lake under the supervision of A. Y. Smith. Those collected in 1969 were analyzed at the Geological Survey of Canada in Ottawa by the author. Forty-two water samples were analyzed for radium in the field laboratory at Bancroft.

Radium and uranium were determined in all sediment and soil samples. Samples collected in 1968 were analyzed for radium by the writer at Queen's University. The remainder of the radium determinations and all the uranium determinations were made by the writer at the Geological Survey of Canada in Ottawa.

Time limitations precluded a thorough analysis of the distribution of uranium with grain size at different distances from the source. Such an analysis would have determined which grain size shows maximum response at any given distance downstream from the source and which grain size to use at different scales of prospecting. Results of a sieve experiment on a clastic sample collected about a mile downstream from Greyhawk mine (Table 14) show that uranium and radium are enriched in fine fractions; therefore, the minus 80 (Tyler) mesh fraction was used for uranium analyses. Radium analyses require a larger sample (about 50 gm), and a coarser fraction (minus 10 mesh) was used.

URANIUM

Uranium determinations were carried out by fluorometry using the method described by Smith and Lynch (1969).

Solid samples, that is soil and stream sediment samples, are leached in hot 4N nitric acid for 2 hours. An aliquot of the leach solution is diluted with 4N nitric acid to give the working sample solution. An aliquot of this solution, equivalent to a sample weight of 5 mg is evaporated on a platinum dish. After a quick ashing of the sample dish at red heat to destroy organic matter, three grams of carbonate-fluoride flux are added and fused in a muffle furnace at 650° C for 10 minutes. Samples are cooled in a desiccator for twenty minutes, and the fluorescence read on a Galvanek-Morrison Fluorometer. Readings are calibrated in terms of uranium content by comparison with standard curves prepared from uranium solutions of known concentration.

A group of 20 samples, selecter over the full range of observed concentrations, were analyzed twice each. Results are presented in Table 7. The standard deviation calculated according to Appendix II is 23%. The significance of analytical and sampling errors is discussed in Chapter 5. Smith and Lynch (1969) cite a detection limit of 0.5 ppm.

Four organic samples were ashed for 20 hours at 500° C and then dissolved with hydrofluoric, nitric and perchloric acids. Uranium determinations on the raw sample, the ashed sample and the dissolved sample are presented in Table 13. It is clear the the simpler method, analyzing the raw sample, detects most of the uranium. This level of accuracy is adequate for geochemical prospecting.

The procedure for water samples is similar to that for sediment and soil. A 5 ml aliquot is evaporated directly on the platinum dish. For samples collected in 1968, the detection limit was 0.1 ppb (Smith and Dyck, 1969). A smaller aliquot (2.5 ml) was used for the samples collected in 1969, and a lower degree of precision resulted. Nine of the 1969 samples, selected over the full range of concentrations, were run twice each and the results are presented in Table 8. The standard deviation calculated according to Appendix II is 49%.

TABLE 7

Replicate Determinations of Uranium in Sediment and Soil

Sample	Number	Uranium	, ppm
31D16	3160	54	46
31F4	7047*	цЦ	34
31F4	7057 *	30	30
31013	3154	19	24
31F4	3843.	9.2	11.8
31E1	6161	7.2	8.6
31D16	3178	6.8	6.6
3114	3679	4.2	5.2
31E1	3178	4.0	3.2
31013	3182	3.0	3.0
31D16	7024 *	1.3	1.0
31F4	373.0	1.1	1.0
31F4	3723	0.8	0.7
31F4	7073*	0.4	0.8
31F4	7034*	0.5	0.7
31D16	6049	0.5	0.6
31F4	7065*	0.7	0.4
31F4	3762	0.4	0.4
31D16	7078*	0.4	0.3
31F4	5029	0.5	0.2

Standard deviation = 23%

*Soil sample

TABLE 8

Replicate Determinations of Uranium in Water

Sample Number	Uraniu	m, ppb
31D16 4151	11	12
31E1 4153	10	11
31E1 4056	3.3	2.8
31D16 4154	1.3	.6
31016 4155	1.0	.3
3114 4152	.9	1,2
31E1 4083	•3	.9
31013 4016	•3	. ધ
31013 4037	.4	.1

Standard deviation = 49%

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RADON

Radon-222 in water is determined by passing air bubbles through the water to collect radon and then measuring the alpha activity of the gas mixture in a scintillometer. The method and apparatus used were developed by Willy Dyck. The following summary of the method and brief description of the apparatus is based on numerous personal communications with Willy Dyck, on his publication (Dyck, 1969a) and on the writer's own experience. For a complete discussion, the reader is referred to Dyck (1969a).

The apparatus consists of five components (see Figure 2): (1) vacuum line with pump and gauge; (2) radon extraction tube fitted at the base with coarse fritted glass disc; (3) radon cell (This is an airtight cylinder, 2.0 inches in diameter and 3.0 inches long, fitted at one end with a glass window and at the other with a "quick-connector." The inside is coated with silver activated zinc sulfide.); (4) photomultiplier tube; (5) electronic counter.

The water sample, 130 ml, is placed in the extraction tube, and the valve to the vacuum line closed. The radon cell is attached to the vacuum line and evacuated. The valve to the pump is closed, and the valve to the extraction tube opened. Air is admitted slowly to the extraction tube through the fritted disc until the pressure reaches atmospheric. At this point virtually all of the radon is in the cell, and it is disconnected.

Ten minutes after the first air was admitted, the cell is placed on the photomultiplier tube and counted for two consecutive five minute intervals. Inside the cell, a pulse of light is given off when each

alpha particle strikes the zinc sulfide. These pulses are then converted to electricity, amplified by the photomultiplier tube and counted by the counter.

The apparatus is calibrated with a 250 pc/1* radium solution from which the radon has been removed and allowed to grow in again according to Equation 3, Appendix I. By letting the standard equilibrate for varying times, one can obtain radon concentrations ranging up to 250 pc/1. The results of the calibrations are expressed as a cell constant in terms of counts per minute (cpm) per pc/1. The cell constant for Dyck's apparatus is 0.333. The amount of radon in a sample in pc/1 is obtained by dividing the cpm by the cell constant.

Radioactive decay must be corrected for unless the analyses are done within a few hours of collection. This is done by dividing the above result in pc/l by $e^{-\lambda t}$ (Equation 1, Appendix I), where λ is the decay constant for radon-222, t is the decaying time and e is the base of the system of natural logarithms, 2.718. Because of the volatility of radon, the sample must be kept in a tightly sealed glass bottle until analyzed.

Other isotopes of radon from the thorium and uranium-235 decay series do not affect the count because of their short half-lives. Radio-activity from other sources, for example, potassium-40, does not interfere either.

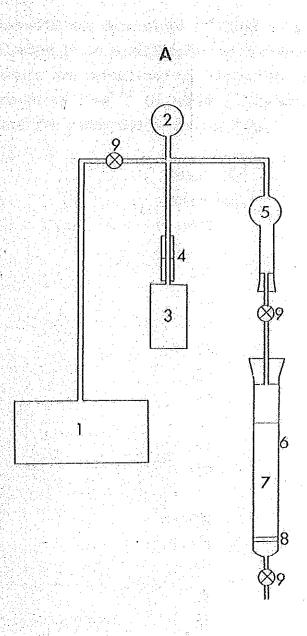
Analytical precision is determined by repeated analyses of the calibrating standard. Seventeen analyses, over a range of radon concentrations from 42 to 240 pc/l, gave a standard deviation of 12% (Table 9).

^{*}A curie is the quantity of a radioactive nuclide in which 3.7×10^{10} disintegrations occur per second or approximately one gram of ${\rm Ra}^{226}$. A picocurie (pc) is 10^{-12} curie.

FIGURE 2

Radon Extraction Apparatus

- A. Complete assembly for extracting radon from water (after Dyck, 1969a)
 - 1. Vacuum pump
 - 2. Vacuum guage
 - 3. Zinc sulfide cell
 - 4. "Quick connector"
 - 5. Drying column
 - 6. Radon extraction tube
 - 7. Water sample
 - 8. Fritted glass disc
 - 9. Valves
- B. Attachment for extracting radon from sediment and soil
 - 10. Water
 - 11. Gas dispersion tube
 - 12. Fritted glass
 - 13. Sediment or soil sample



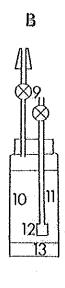


TABLE 9

Replicate Determinations of Radon in Water

Radon-222 was determined 17 times in a 250 pc/l radium-226 standard by allowing it to equilibrate for different lengths of time. For comparison, results are normalized to redium-226 equivalent by dividing the radon-222 content by $1-e^{-\lambda t}$ (Equation 3, Appendix I), where λ is the decay constant for radon and t is the time.

	Radon-222	Radium-226
	detected,	equivalent
	pc/1	pc/1
Mary Co. See Gero	en e	
ling. Nigovana – Kolonova	135	265
	110	265
The three sectors	37	222
	65	219
	158	239
	62	225
	55	1.98
	51	267
	118	198
	77	279
	135	220
	. 204,	204
The state of the s	115	176
		192
	44. 1 4 37 446. 41	222
INDER OF STATE		261
	221	268
		r News

Standard deviation = 12%

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This precision is not strictly comparable to that of the analytical methods for radium and uranium because the range of concentrations used was limited to the upper part of the natural distribution.

The largest source of error, except in the case of high samples, is the statistical error of count rates. For a counting time of 15 minutes (5 minutes background and 10 minutes sample) the detection limit is 1 or 2 pc/1. At this level of sensitivity, the output per assembly is about 27 samples per eight-hour day. One person can, however, operate more than one assembly.

Application of the method is limited by the half-life of radon, 3.82 days. In 15 days, a 32 pc/l sample (fairly high) would decay to 2 pc/l. Analyses must therefore be carried out within a few days of collection. This requires that the apparatus be set up in the field. Sending the samples to a commercial laboratory would be feasible only in rare cases.

RADIUM

The classical method for determining radium-226 (for example, see Faul, 1954, pp. 42-48) consists of letting nascent radon-222 form from radium-226 according to Equation 3 (Appendix I) and then measuring the nascent radon. Radium-226 in liquid is determined by a simple modification of Dyck's (1969a) method. Solids, in the classical method, are dissolved or fused to release the radon. Development of a simpler method for determining radium-226 in sediment and soil, described in the remaining part of this chapter, is a major contribution of this thesis.

Equation 3 (Appendix I) states that radon-222 will grow into equilibrium with radium-226 with a half-life of 3.82 days. Thus the time limitation for radium determinations is exactly opposite to that of radon: the sample must sit for a minimum of a few days rather than a maximum.

RADIUM DETERMINATIONS IN WATER

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Radium-226 in water is determined in the same way as radon except for two changes. (1) All the radon present initially in the water is removed by flushing with air bubbles or by allowing it to decay. The sample is then allowed to sit for several days for fresh radon to grow in. (2) Radium-226 in pc/1 is equal to

cpm
$$\frac{}{\text{cell constant} \times (1 - e^{-\lambda t})}$$

where λ is the decay constant for radon-222 and t is the equilibrating time (Equation 3, Appendix I). Productivity, precision and detection limit are the same as for radon.

RADIUM DETERMINATIONS IN SELIMENT AND SOIL

Dissolving or fusing a sediment or soil, as required by the classical analytical method for radium-226, is a lengthy procedure and unsatisfactory for geochemical prospecting which requires cheap

analyses. In geochemical prospecting methods, dissolving is usually replaced by leaching with water or acid, or by partial fusion with a flux such as potassium bisulfate. These processes are faster but are less complete in freeing the element of interest. A major purpose of this thesis is to develop an analytical method for radium-226 in sediment and soil which is simple enough and of sufficient precision and accuracy to be of use in geochemical prospecting.

In the case of radium determinations, only the radon-222 must be removed from the solid, because it is radon which is measured. Radium-226 on or near grain surfaces gives up its radon spontaneously. Radium inside large grains is not detected, but it is of less value to geochemical prospecting than is adsorbed radium. Adsorbed radium travelled to its present location in solution in the water, and this water can usually be traced upstream to its source. The distribution of large grains with enclosed radium, on the other hand, was affected more by glaciation, and their source is less obvious.

Rapid methods of radium-226 determination developed in this thesis can be divided into two types depending on the medium, air or water, in which nascent radon is stored as it is growing in. In this work, a water medium was used for routine determinations, because bottles were available to the author whereas airtight vessels must be manufactured in large numbers. However, some pilot tests were made using air, and this method was found to be simpler and more precise than those using water (Table 16).

Methods Using Water as a Storage Medium

Two methods were tried: the first gave erratic results; the sec-

ond gave better reproducibility and was used in routine determinations. The results of replicate analyses of several samples using the two methods are presented in Table 10.

Method 1: In the first method, about 50 grams of sediment are put into a 260 ml glass bottle which is then filled with water free of radon and radium. This is shaken and allowed to sit for several days. The length of time allowed depends on the sensitivity required. Little is gained after ten days. The water is then decanted into the radon extraction tube and radon-222 in the water is measured by the method previously described above (page 35).

Radium in the sediment in pc/gm should then be equal to

$$\frac{\text{cpm} \times (.260 - V)}{\text{cell constant} \times (1 - e^{-\lambda t}) \times W},$$

where V and V are the volume in liters and the weight in grams respectively of the sediment, λ is the decay constant for radon-222 and t is the equilibrating time. The expression 0.260-V is the volume of water in the bottle. The formula assumes that all the radon gets into the water. This is unlikely because the sample is left undisturbed for several days. The alternative is to shake the bottle again just before decanting. This is of only moderate help because there is always an air bubble in the bottle which absorbs some of the radon during shaking and which, when the lid is opened, is lost.

Method 2: In the second method, which is more precise, the water

TABLE 10

Comparison of Results Obtained by Two Radium Methods with Radium Expressed in pc/gm

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Sample Number Method 1	Method 2
31D16 3163 75.1	337
110	240
	236
131 57.7	257
84.5	
31F4 3409 1.81	6.60
2,82	5.29
1.86	5.27
1.25	4.52
31.013 31.61 26.8 76.1	117.5
98.4	
31013 3160 5 69 7 60	17.4
7.60 10.1	,

The state of the s

is bubbled in the bottle by letting air in through a glass gas dispersion tube. Because this was the method used in routine analyses, the detailed procedure is given in Appendix III, and a brief description of the method follows.

After about 50 grams of sample has been allowed to sit in the bottle filled with water for several days, the lid is removed and 50 ml of water poured out and discarded. Then a two-hole rubber stopper is attached (see Figure 2). One hole contains the gas dispersion tube which reaches to about the top of the sediment. The other contains a short glass tube which reaches only the bottom of the stopper and is connected, at the top, to the drying column. Both of these tubes have stop-cocks. These are closed and the bottle shaken. The cell is evacuated, and the radon extracted by admitting air as a multitude of tiny bubbles through the gas dispersion tube.

The greater precision of Method 2 relative to Method 1 is apparently due to the fact that in Method 2 the sediment and water can be shaken vigorously without radon being lost to the bubble. Some radon is lost when the 50 ml of water is discarded, but this can be corrected for roughly in the method of calculation.

Using one assembly, the author was able to run 20 to 25 samples per eight-hour day. One person could, however, run two or three units at once for an output of over 50 samples per day. Output per assembly could be increased by reducing the counting time with a sacrifice of precision and detection limit.

Calibration: The apparatus was calibrated using a 250 pc/l radium-226 standard, and a new "cell constant" was arrived at, this time in terms of cpm/pc. Pouring out 50 ml of standard reduces the volume to 210 ml. The bottle then contains $0.210 \times 250(1-e^{-\lambda t})$ pc of radon, where λ is the decay constant for radon-222 and t is the time that the standard was allowed to equilibrate. The cell constant is then

$$\frac{\text{cpm}}{.210 \times 250(1 - e^{-\lambda t})}$$
.

Values of this cell constant and of the number of counts observed for seven calibration runs are presented in Table 11. The 95% confidence limits for the cell constant are 1.22 ± 0.16 (obtained from the Student -t distribution with six degrees of freedom).

The standard deviation of the cell constant is 0.175 or 14%. The average of the number of counts obtained is 478. The standard error of count rates is equal to the square root of the number of counts, in this case 21.9 or 4.6%. The disparity between these two error estimates indicates that the experimental error in determining the cell constant is considerably in excess of that due to low count rates. It is also greater than that reported by Dyck (1969à). His 26 determinations of the cell constant have a standard deviation of 8%. Because the rest of the method was identical to Dyck's, the source of the error must lie in the method of bubbling.

Radon extraction efficiency: Dyck (1969a) reports that the extraction efficiency of his radon extraction tube is 95%. The efficiency using the gas dispersion tube, used in this study, is lower, about 50%. This figure is arrived at in two ways.

TABLE 11

Determination of Cell Constant for Radon System Using Gas Dispersion Tube

	Counts	Cell Constant
		cpm/pc
	44	
	540	1.09
	270	1.11
	447	1.00
•	671	1.29
	558	1.01
	179	1.18
	678	1.46
Average	478	1.22
Standard Error	21.9	.175
Relative Standard Erro	r 4.6%	14%

TABLE 12

Radon Extraction Efficiency Using Gas Dispersion Tube

ting recognition, allowing a literature of the principle of

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cpm run l	cpm run 2	cpm run 2 cpm run 1	cpm run 3	cpm run 3 cpm run 2
	प्रकार <u>भूकी</u> देखनाते । -			-
73.7	34.3	.466		
190.8	88	.461		
203.9	84.6	. 434	35.1	.416
1462	568.6	. 389	·	
	week a second of	and the Administration		

- (1) A series of samples was tested 2 to 3 times each in immediate succession, allowing no time for radon to build up (see Table 12). The fraction of radon remaining in the water is the cpm divided by the cpm of the immediately preceding run. The efficiency is one minus this ratio. The average efficiency of five pairs was 57%.
- (2) The cell constant using the gas dispersion tube is compared with that reported by Dyck (1969a). He gives a value of 0.333 cpm per pc/1 or, because he used 130 ml of standard, $.333 \div .130 = 2.56$ cpm per pc. Because this was based on an efficiency of 95%, 100% of the radon would give $2.56 \div .95 = 2.70$ cpm per pc. The efficiency of the gas dispersion tube is then $1.22 \div 2.70 = 45\%$

Radon extraction with the gas dispersion tube is both less precise and less thorough than with the radon extraction tube. In spite of the inferior precision of the gas dispersion tube applied to a standard radium solution, sediment and soil samples can be analyzed for radium with greater precision using the gas dispersion tube than using the radon extraction tube.

Calculation of radium: In calculating the amount of radium in the sediment, account should be taken of the reduction in volume of water by the volume of sediment. If the porosity of the sediment is assumed to be 50%, then the volume of water available for radon storage is $260 - \frac{V}{2}$, where V is the volume of the sediment. When the sample is bubbled, the water trapped within the sediment does not circulate and hence does not give up its radon. A further 50 ml of water is discarded. The volume of water tested is then 210 - V. Thus the fraction measured of the total radon given up by the sediment is equal to

$$\frac{210 - V}{260 - \frac{V}{2}}$$

The amount of radium in the sediment or soil tested is then

$$\begin{array}{ccc}
\text{cpm} & 260 - \frac{V}{2} \\
1.22V(1 - e^{-\lambda t}) & 210 - V
\end{array}$$

in pc/gm, where W is the weight of sample used. Derivation of the cell constant, 1.22, was explained on page 46. Because some radon is trapped in the sediment when the 50 ml of water is discarded and then is released when the bottle is shaken, the discarded water contains less than the "average" amount of radon, and this calculation will give an answer which is slightly too high. In the extreme case where no radon moves from the sediment into the water before the sample is shaken, the radium concentration as calculated in this section will be a factor of

$$260 - \frac{v}{2}$$

$$210 - \frac{v}{2}$$

too high, that is about 25% too high. A maximum analytical bias of this magnitude is not serious in geochemical prospecting. Precision is more important than accuracy, and Method 2 was the most precise method available to the author.

The above calculation can be simplified by assuming that V is

the same for every sample. With V equal to 40 ml, the formula reduces to

$$\frac{1.16 \text{ cpm}}{\sqrt{(1 - e^{-\lambda t})}}$$

In a series of replicate analyses, where 22 samples were analyzed 47 times, the decrease in precision resulting from this simplification was slight (standard deviation, S = 32% compared to S = 31%), even though V ranged from 8 to 40 ml. Samples collected in 1968 were calculated by the simpler method, assuming V = 40 ml. Those collected in 1969 were calculated by both methods, but only the results of the longer method are used in this thesis.

Another possibility is to calculate the radium in parts per volume or pc/ml. This is accomplished by substituting V for W in the above formula. For the same set of replicates, this again led to a slight decrease in precision (S = 33% compared to S = 31%).

Interference: Sources of interference are limited to alpharadioactive gases originating in the samples. Other sources of alpharadiation, such as impurities in the apparatus or laboratory air, are controlled by checking the background.

Except for the U²³⁸, U²³⁵ and thorium decay series, natural alpha-radioactivity is limited to isotopes of cerium, neodymiun, samarium, gadolinium, hafnium and platinum. These nuclides are too long-lived and of too low abundance to make a significant addition to the alpha-radio-activity of radon. Moreover, none of them are volatile. In addition,

radon has several solid daughter products which contribute alpha-radioactivity in the cell.

Sources of interference are limited therefore to members of the U^{238} , U^{235} and thorium decay series. Each of these three series contains an isotope of radon (Tables 1, 2, and 3). None of the other members are volatile.

In natural uranium at equilibrium, Rn²²² is about 25 times as active as Rn²¹⁹. Furthermore, the half life of Rn²¹⁹ is so short (3.92 seconds) that virtually all of it decays after it leaves the sample before it is counted.

Interference from Rn²²⁰ (thoron) from the thorium series is both more likely and, from the point of view of geochemical prospecting, more important. Thoron has a half-life of 52 seconds and has no long-lived alpha-radioactive daughters. Interference from thoron can thus be prevented with judicious timing. A period of 10 minutes elapses between shaking the sample (releasing the short-lived thoron to the water from its solid parent Ra²²⁴ in the sediment) and beginning the count. During this 10 minutes virtually all the thoron will decay.

The minimum level of thoron which will give a measurable count can be calculated. Dyck (1969a, p. 14) states that thoron gives a count rate of 2 cpm/pc one minute after isolation from its parent. In the interval, 10 - 20 minutes after isolation, thoron will give a total count of

$$\frac{2}{60} \int_{540}^{1140} e^{-\lambda t} dt = -\frac{2}{60\lambda} e^{-\lambda t}$$
 counts per pc

or, because $\lambda=0.0127~{\rm sec}^{-1}$, a total count of 3×10^{-3} counts per pc. Dividing by the number of minutes counted gives a count rate of 3×10^{-4} cpm/pc. The radon-222 counting rate for the same period is 2.56 cpm/pc (see page). Thus, in terms of radioactivity, it takes 8500 times as much thoron as radon to produce the same count. In terms of mass of ${\rm Th}^{2.32}$ and ${\rm U}^{2.38}$ in equilibrium, the same ratio is 26,000. The thorium-uranium ratio must, therefore, be well in excess of 1000 in order to have a significant effect on radium determinations. The ratio in igneous rocks is fairly constant at 3.5, and such an enrichment is extremely unlikely in the Bancroft area.

A further control on interference by thoron is provided by counting for two five-minute intervals instead of one ten-minute interval.

Interference by thoron will be evinced by a sharp drop in count rate.

Monitoring the two count rates during the course of the routine analyses showed no interference from thoron.

Accuracy: The accuracy of the method was tested by running eleven sediment samples with a standard method. The samples were dissolved with hydrofluoric, nitric and perchloric acids. The radon was allowed to grow in and then was measured as previously described, except that a 60 minute counting time was used for greater precision.

Four organic samples were run. They were each sieved to -25 mesh and then split into three portions. One portion was analyzed in the routine manner by Method 2 above. The other two were dry ashed for 20 hours at 500° C. The ash was again split into two portions: one portion was analyzed by Method 2, and the other (1 gm) was dissolved. Results are expressed in pc/gm in Table 13. A correction has been made

TABLE 13

Comparison of Analyses of Untreated, Ashed and Dissolved Fractions of Organic Samples

ll U/Ra Dissolved		57	10		88	43		ci Ci	(V)		7.7	4.7
10 U/Ra Ashed		77			55	汉		∞ ~i	, i		ر. ال	919
9 10 U/Ra U/Ra Not Ashed Ashed	% .9			IJ			1 10			2.7		
7 8 U Ashed U Dissol- 1 ppm ved ppm 1		12.1	14.5		& 9	rd G		φ. ær	ω		80	۳,
7 U Ashed ppm		14.6	12.4		<u>ئ</u>	± ∞		iņ W	ω at		37	88
6 U Not Ashed ppm	10.4			\$			0.0			70		
5 Ra Dissol- ved pc/gm		. 82	7		∞.	7.7		m	ന്		9	11.5
ų Ra Ashed pc/gm		.87	7,06		.95	1.55		3.0	Ω ω		74.6	2,0
3 Weight Ra Not Loss % Ashed pc/gm	1.52			2.7			1.32			74.8		
2 Weight Loss %		27	1.2		35.4	35.7		27.1	26.0		ω	.√ ∞
l Sample Number	31E1 3155	ASD 1	Ash 2	31F4 3793	Ash 1 35.4	Ash 2 35.7	31月4 3835	Ash 1	Ash 2	3174 3410	Ash 1	Ash 2

for weight loss on ashing so that results in columns 3, 4, and 5 should be the same for a given sample. The discrepancy in the weight loss between the two ashed fractions of sample 31E1 3155 cannot be explained.

A similar test was made on clastic material. Sample 31F4 5169 was sieved into seven fractions, and each fraction was analyzed both by the routine Method 2 and by dissolving. Dissolving was carried out as before except that the samples were not ashed. The coarse fractions were ground before being dissolved but not before the routine determination. Results are expressed as pc/gm in Table 14.

The results are encouraging. The routine method detects most of the total radium. A level of accuracy is indicated that is adequate for geochemical prospecting.

A surprising result shown by Table 14 for the four finest fractions is that the routine method gave a higher value than the more thorough determination on dissolved material. The two methods are still close, however, if one excepts the finest fraction. The most likely explanation is that some radon has accumulated in the fine, dry sediments and was not removed when the samples were weighed into bottles. The method of calculation assumes that all such radon is lost. Radon is apparently freed more readily when the sample is submerged in water and shaken than when it is dry. Further evidence of this is given on page 64.

The fraction detected by the routine method increases with decreasing grain size. Either of two explanations is possible. (1) Radon which had accumulated in dry sediments was, as would be expected, more thoroughly removed from coarse fractions than from fine before water was added. (2) Some radium is far enough inside the grains that radon

TABLE 14

and Dissolved Splits of Different Size Fractions of Clastic Sediment of Analyses

5169	
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Number	
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son of Analy		e i grande grande <mark>ge</mark> Geografia	Size	Fraction			9 +	10 1 20	20 - 60 - 60	90 - 80	8	150 - 200	-200	Total
Ses of		० हाइन कर	Weight	(A)			77.5	283	105 107	85.00 0.00	27.3	7.	7.00	821
Intreated	en gran	ന	% O	Total	Weight		or or	ų ų	49.0	77.8	2.60	Ŋ	₩,	
or or		anily mag	Ra Mot	Dissolved	pc/gm		9.	2.0 1.6	2.2					
ved Spli	e Number	ĸ	⇒ x w							. 198	7	070	189	2.57 pe/em
ts of Differ	3184 5169	ω	සූ	Dissolved	pc/gm		্ৰ পে	٠ .	m m	iņ Vi	-	12.1	15.7	
rent Size		[~~	m x v				8.	д .3%	7.62	.163	. 193	.062	. H	3.87 pc/gm
Fractions of		ω	Þ	Dissolved	mad		d ⊲.	H		ı 	۷. ص	ເບ	6.9	
		O	U/Ra				.35	82.		[2.	æ.	3	7	
	and Dissolved Splits of Different Size Fractions of Clastic	and Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169	'Q	and Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 4 5 6 7 8 9 Ra Not 4 x 3 Ra 6 x 3 U U/Ra	and Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 Ra Not 4 × 3	and Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 Ra Not 4×3 Ra 6×3 U U/Ra Dissolved Dissolved Dissolved Dissolved ppm	and Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 4 5 6 7 8 9 Fa Not 4 × 3 Ra 6 × 3 U U/Ra Dissolved Dissolved Dissolved po/gm ppm	and Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 Ra Not 4×3 Ra 6×3 U U/Ra Dissolved Dissolved Dissolved Dissolved Dissolved Spm	and Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 Ra Not 4 × 3 Ra 6 × 3 U U/Ra Dissolved pc/gm pc/gm pc/gm ppm 2.6 .24 3.4 .32 1.2 .35 2.0 1.6 .62 4.0 1.38 1.1 .28	And Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 Rangle Number 31F4 5169 Rangle Number 31F4 5169 Rangle Number 31F4 5169 Paragle Number	A Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 Ra Not 4 × 3 Ra 6 × 3 U U/Ra Dissolved Dissolved Dissolved Ppm Pc/gm Pc/gm Ppm Pc/gm	And Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 Fa Not 4 × 3 Fa 6 × 3 U U/Fa Dissolved Dissolved Ppm Pc/gm	And Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 Ra Not 4 × 3 Ra 6 × 3 U U/Ra Dissolved Dissolved Dissolved Dissolved Dissolved 2.6 .24 3.4 .32 1.2 .35 2.0 1.6 .62 4.0 1.38 1.1 .28 1.8 2.2 .98 3.3 1.62 6.3 .198 5.2 .163 1.1 .21 10.4 .271 7.4 .193 2.9 .39 13.7 .070 12.1 .062 5.5 .45	and Dissolved Splits of Different Size Fractions of Clastic Sample Number 31F4 5169 Ramble Number 31F4 5169 Park Not 4 × 3 Fa 6 × 3 U U/Fa Dissolved po/gm ppm ppm 2.6 .24 3.4 .32 1.2 .35 2.0 1.6 .62 4.0 1.38 1.1 .28 2.0 1.6 .62 4.0 1.38 1.1 .28 2.0 1.6 .62 4.0 1.38 1.1 .28 2.0 1.6 .62 4.0 1.38 1.1 .28 2.0 1.6 .62 4.0 1.38 1.1 .28 2.0 1.6 .62 4.0 1.38 1.1 .28 2.0 1.6 .62 4.0 1.38 1.1 .28 2.0 1.6 .62 4.0 1.38 1.1 .28 2.1 1.9

doesn't get out, the fraction trapped in this way being higher for coarse grains than for fine.

Precision and detection limit: Comparison of the precision of Methods 1 and 2 can be made by calculating the standard deviation S as described in Appendix III. The values used are those in Table 10. Mehtod 1 gives S = 36% if all the samples are used, and 31% if only those on which replicate analyses were made by Mehtod 2 are used. Method 2 gives S = 16%. In addition to better precision, Method 2 recovers and detects a larger amount of radon.

Because these tests showed that Method 2, the method using the gas dispersion tube, gave better precision, it was used for routine analyses. During the course of routine analyses, 44 samples were run twice each. These were treated in exactly the same way as the other samples and were selected over the full range of radium values. In many cases the weight of sample used and the length of equilibrating time used in each determination was varied.

Results of the replicate analyses are presented in Table 15. A standard deviation of 33% resulted from the 44 replicate determinations. The standard deviation given above, 16%, is lower because in that case samples containing larger amounts of radium were used, and these samples give lower relative errors.

A large part of this error is due to the randomness of radioactive decay. For high samples this error is unimportant, but for low ones it is the most important source of error. Another source of error is the inconsistency of transfer of radon from water to air in the bubbling procedure. The importance of this was demonstrated on page 46.

TABLE 15

Replicate Determinations of Radium in Sediment and Soil Using the Routine Method (Method 2)

Sample Number	Radium	pc/gm	÷ 1	Sample	Number	Radium,	pc/gm
31D16 3163	170	180		31E1	31.75	,60	.72
31013 3154	54	40		31F4	3790	.62	.43
31016 3160	29	59	Mark Tolk	31E1	5160	. 46	.53
31E1 5001	18	21.		31C13	3150	. 15	.80
31D16 7016*	19	15		31F4	5166	. 44	. 49
31E1 5005	12	10		31F4	3695	.43	.46
31D16 3180	9.0	11		31E1	5165	. 30	•55
31E1 5004	7.2	9.2	•	31F4	5167	.48	.30
31F4 3409	3.7	3.2		31E1	5161	. 44	,19
31E1 5092	3.3	2.7		31013	3158	.19	.27
31D16 3182	2.5	3/3	the Land	31F4	3666	,26	.18
31174 6137	2.2	3.2		31F4	3723	.25	.17
31E1 5164	2.7	2.1		31F4	5168	.26	.11
3154 5169	2.3	2.4		31D16	5051	.07	.20
31E1 6165	1.8	2.7		31F4	5022	.10	.16
31D16 7019*	1.7	2.7	•	31F4	3767	.10	. 14
31E1 5162	2.4	1.8		31013	3171	.12	.11
31E1 5163	1.2	2.7		31C13	3189	.10	.06
31F4 7036*	2.0	1.5		31k/i	3719	.06	.04
31F4 3848	1.0	1.2		31F4	3788	.04	.06
31F4 3817	.89	1.0		31013	3148	.04	.06
31E1 3153	.87	.48		31)74	3693	.05	.02
	to war and the		, i				
A Committee of the Comm							

Standard deviation = 33%

#Soil sample

Inconsistency of transfer of radon from sediment to water is a further probable source of error. Manipulation errors, errors in weighing, etc., are unimportant at this level of precision. Electronic difficulties are not serious (Dyck, 1969a).

A check was made to determine whether the incomplete removal of radon from the dry sample had an adverse effect on the analytical precision. In 21 of the replicate pairs discussed on page 57, the equilibrium time was different for the two members. If incomplete removal of radon is important, then longer equilibration times will result in lower radium values. In 12 of the 21 pairs this resulted, in 7 the reverse was observed, and in three there was no change. The results are inconclusive and could well be fortuitous. On the other hand, retention of radon may have a small adverse effect on precision.

Sensitivity is limited by the error of low count rates, the background of the cell and the amount of material available. During the course of the analytical work, background increased from 0.1 cpm quoted by Dyck (1969a) to an average of about 0.5 cpm. Undoubtedly this was due to the accumulation of solid radioactive decay products on the cell walls. Samples with net count rates lower than this will give unreliable results. The amount of sample used was about 25 gm. If a 25 gm sample is left for ten days and gives a net rate of 0.5 cpm, its radium content is about 0.02 pc/gm. This then is the detection limit. Earlier this writer (1969a) reported a detection limit of 0.01 pc/gm. This lower figure was a result of a lower background. The detection limit could be lowered and the precision improved somewhat by counting for a longer period.

Methods Using Air as a Storage Medium

The most exacting and time consuming step in the methods using water as a storage medium is extraction of radon from the water. Most of the imprecision of these methods is due to variations in the efficiency of extraction of radon from water. Some imprecision is probably due to inconsistencies in transfer of radon from the sediment to the water. An improvement in reproducibility, detection limit and productivity can best be achieved by storing the radon in some medium other than water. Air is an obvious choice. Radon is again determined with Dyck's apparatus minus the extraction tube.

Three different types of vessel were tried: glass and rubber, plastic, and brass. Radon disappeared from both the glass and plastic containers. It may have leaked out or been absorbed by the container walls. Sedlet (1966) reports that radon is adsorbed onto glass and diffuses through rubber and plastic. Brass, however, gave good results.

In order to achieve an output of 1000 samples per week, a worth-while output for a commercial laboratory, and still allow one week for equilibration time, 1000 vessels would be needed. The unit cost, therefore, is an important factor in choosing a design. The other requirements are: (1) a valve for letting radon out, (2) a means of connecting this valve to the evacuated radon cell and (3) a means of opening the container to change samples. Efficiency of radon extraction can be improved by installing a second valve to admit fresh air. The radon is then flushed out stepwise by opening and closing each valve alternately.

Two pilot vessels were built, at the Geological Survey of Canada, of 0.36" I. D. brass tubing, about 30 inches long. For routine determinations much shorter tubes could be used, six inches long would be

ample. Each end is threaded on the inside and fitted with a valve. Samples are changed by unscrewing the valve. Cotton is placed between the sediment and the valve for protection.

Original radon is removed by connecting the tube to a vacuum line and flushing several times with air. Radon is then allowed to grow in for several days. A radon cell (Figure 2) is evacuated and connected to a valve. This valve is opened and then closed. Most of the radon is now in the cell. The second valve is opened and closed allowing fresh air to flow into the tube. Repeating this process several times forces nearly all the radon into the cell. The cell is then counted in the normal manner.

Difficulty was experienced in obtaining an airtight seal between the valve and the brass tube. The problem was solved by coating the threads with teflor tape.

Loss of radon is recognized by allowing the sample to equilibrate for different lengths of time and observing whether radon is accumulating according to Equation 3 (Appendix I). The results of such an experiment on sample 31F4 5169 (not sieved) are presented in Table 16. The sample material was not changed during the course of the experiment so that only two fractions of about 65 grams each were used with one placed in each tube.

Table 16 shows that leakage of radon is not important. All the variation in columns 5 and 6 could be explained as due to count error*. A close look at column 5, especially the last item, reveals that values

^{*}Count error percent is equal to $\frac{\sqrt{n}}{n} \times 100$ where n is the number of counts observed. If a sample of constant activity is counted many times, then 68% of the results will lie within the count error percent of the mean.

TABLE 16

Results of Radium Determinations Using Air

. 1	2	3	4	5	6	7
Tube No.	Counts	Count	Time	cpm	5	Ra pc/gm
	Observed	Error %	Hours	$1 - e^{-\lambda t}$	weight	
1	533	4.3	24	313	5.02	1.88
	1846	2.3	125	300	4.82	
	2927	1.8	424	300	4.82	
2	676	3.8	26	376	5.04	1.89
	1431	2.6	65	367	4.92	
2.10	2304	2.1	125	374	5.01	
	6670	1.2	424	353	4.73	

do drop of slightly with time, suggesting a slight radon loss. The numbers in column 6 are proportional to the radium concentration. They show that the radium concentration is the same in both sample fractions, and that radon extraction efficiency is the same in both tubes. The precision of this method is much higher than that of methods using water (Table 10).

In order to relate results to radium concentration, it is necessary to make some assumption about the efficiency of transfer of radon from the sediments to the cell. One hundred percent is a logical first approximation. This can be checked by comparison with the results of the solution experiment (Table 14) and with the results obtained by the routine method.

The average of the values in column 6 divided by the cell constant will give the radium content. The cell constant of Dyck's apparatus is 2.60 cpm/pc (see page 49). Radium content is shown in column 7 and is the same for both tubes.

The best radium value for sample 31F4 5169 is given in Table 14 at the bottom of column 7. This is obtained by weighting the individual dissolved radium values (column 6) by the proportion of the total weight represented by each fraction (column 3) and then summing. The result of the same calculation for the solid determination is shown at the bottom of column 5.

Values of radium for sample 31F4 5169 obtained by four different methods are presented in Table 17. The five routine determinations and the two determinations in air represent seven separate portions of the sample. The sieving experiment was carried out on still another portion. The close agreement between different runs by the same method shows that

sample inhomogeneity is not important. The difference obtained between methods using air (2.0 pc/gm) and using water (2.5 pc/gm) is therefore real.

Choice between alternate analytical methods in geochemical prospecting is made on the basis of reproducibility or precision; simplicity; and thoroughness of extraction and detection, or accuracy. Using air as a radon storage medium is simpler and much more precise than using water. Extraction of radon into air, on the other hand, is slightly less thorough than into water. The difference in thoroughness between the two methods is slight however compared to the difference between them and the "true" value, 3.9 pc/gm, obtained by dissolving. There is little choice between the two methods on the basis of thoroughness. The greater precision and simplicity using air weighs heavily in favor of that method, thus anyone planning to do radium analyses on a commercial scale should give strong consideration to using air. A large number of sample storage containers, probably of brass, would however have to be constructed.

Curiosity is aroused by the fact that more radon was recovered using water as a storage medium than using air. The best explanation is that radon emanation into water is more efficient than into air. Another explanation worth considering is that all the air is not swept from the tube into the cell. A sweeping efficiency of 2.0 ÷ 2.5 = 80% would account for the difference. Such a low efficiency is unlikely in view of the stepwise nature of flushing the tube. Furthermore, Dyck (1969a) reports a radon extraction efficiency from water of 95%, and his procedure does not include stepwise flushing. The conclusion remains that radon enters water more easily than it enters air.

74 mars and the state of the TABLE 17

Radium Determination (pc/gm) in Sample 31F4 5169 by Four Different Methods

Weigh	ted Average	Routine	Determination
of Si	eved Sample	Determination	in Air
(Ta	ble 14)	(Method 2)	(Table 16)
Dissolved	Not dissolved		
3.87	2.57	2.34	1.98
		2.43	1.99
		2.40	
		2.33	
		2.56	
Av	erage	2.41	1.99

Vinogradov (1959, p. 165) comments: "[In] moist sands...radon diffuses easily and is readily extracted by circulating water. Well drained soils are poor in radon. Dry soils are relatively more radio-active." In other words, radon is removed more efficiently from wet soils than from dry. Evidently the same process is active in the laboratory as in the field.

CHAPTER 5 STATISTICAL PRESENTATION OF RESULTS

MEANS, RANGES, ETC.

Results of routine analyses are presented in Table 18 and Figures 3, 4 and 5. Drainage samples clustered around mines or in drainage which may have been contaminated were omitted. Data for organic sediments include data for plant parts. For uranium in water only samples collected in 1968, 76 in all, were included in the calculation of median and range. Those collected in 1969 were analyzed with less precision, and most were below the detection limit of 0.5 ppb. Only 42 water samples were analyzed for radium, and results were too low to be of use.

Some results are worthy of note at this point. Both uranium and radium are enriched in organic relative to clastic sediments (see Chapter 1). Uranium and radium levels in samples containing plant parts are intermediate between those for clastic and organic samples.

Radon, on the average, is low in lakes and ponds, intermediate in streams and swamps and high in springs (Figure 3 and Table 18). Low levels of radon in lakes and ponds are believed due to the depth of these bodies and the resulting high ratio of water to sediment, the major source of radon (see Chapter 9). High levels of radon in springs have two causes. (1) Groundwater is in more intimate contact with (radiumbearing) solids than is surface water. The area of rock or sand in contact with a given volume of groundwater is much greater than the area of sediment in contact with the same volume of surface water. Thus, the ratio of radium to water is higher for groundwater than for surface water. (2) Radon is less likely to escape from groundwater due to the

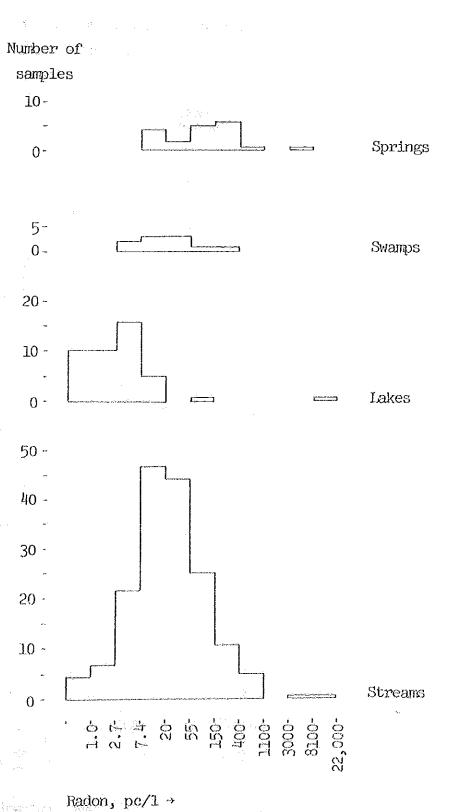
TABLE 18

Statistical Presentation of Results

		•				Samp Erroi Space	7 % I	Analytical Error %	Approximate Detection Limit
			No.	Median	Range				
) 			3 (0	.92	,02- ¹ 100	21	<u> </u>		
	OL	Organic Sediment		.92	.07-12	.	[31	.02 pc/gm
	_	Plant Parts	37		.07-12	1	38	٠.٠	
ants	ස <u>ූ</u>	Clastic Sediment	155				30)		
Jim		Clastic Sediment Organic Sediment Plant Parts	162	3.4	<220	48		-	0.5
Se	HOO	Plant Parts	37	2.6	0.6-170		}	23	0.5 ppm
	ב	Clastic Sediment	153	1.3	<92	30	50		
		Streams	173	23	<12,000	7			
	d	Lakes and Ponds	43	3	<9000	(65.69	7.0	1 20/1
	ပ္သ	'Swamps	10	20	4-150	14	57	12	1 pc/1
	E	Springs	19	120	10-3350)		gang J. Ladaban 1997, Apid 1997 was and S. Wijahayan was to dog Tabahan sawar	agi ka diangganggang Kanlaga da kala da diang menjalangga pada ngan 1 sa Salanda ngan ang menjanggan an
2.2		Streams	48	0.2	<110)			
ate	q	Lakes and Swamps	26	0.1	<12	67	117	49	.l ppb
3		Springs	2		<4.4)			
	-	Streams	19	0.9	<2.8)			
	· ` `	Lakes and Swamps	18	0.9	<2.4	}			1 pc/1
		Springs	5	0.4	<2.1)		an alaka sejalah ali seperji Maliferbolik da Alikasan semperangan palamingan da Sejalah kelalah da	
	1	A horizon	40	12	1.5-180	12	1		00 11 /
	S C	B horizon	36		.27-65	69.	\	21	.02 pc/gm
뜅	x	A horizon	40	3	<500	73)	والمناسبة فالمواودة والمستوفي والمواودة بيام فالموامث والموامث والموامث والمستحدود والمتحدود	in fact the second of Philosophic Control and an interference and design and a second control control control of
1 13		A norizon B horizon	36		<150	27	}	23	0.5 ppm

FIGURE 3

Distribution of Radon in Surface Water



PIGURE 4

Distribution of Radium and Uranium in Sediments

(Asymmetry of uranium histograms is due to lack of sensitivity of analytical method at low levels.)

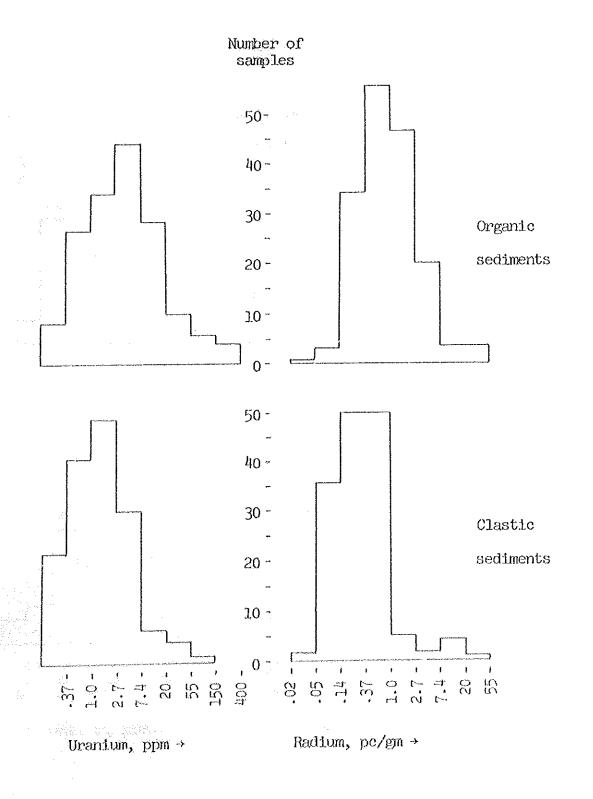
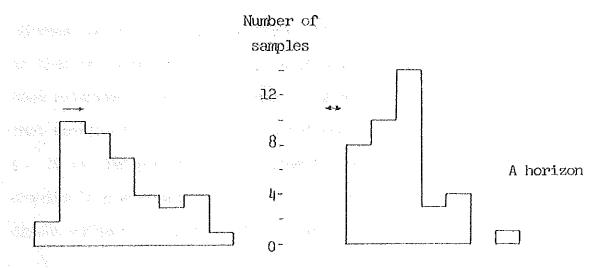


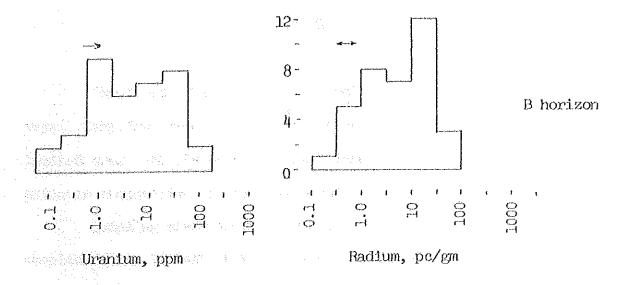
FIGURE 5

Distribution of Radium and Uranium in Soil

Arrows indicate background as explained in Chapter 9.

Bap vala da agrecieto e filoso e e e e e e





absence of air.

Population distributions for radium and uranium in sediment and radon in water are approximately lognormal (Figures 3 and 4). This is the usual case for trace elements (see Hawkes and Webb, 1962, pp. 25-26). The same is apparently true for radium and uranium in soil although not enough samples were collected to be sure (Figure 5).

Radium is enriched in the A horizon of soils relative to the B, whereas the opposite is true for uranium. The more likely explanation is that the A horizon is the zone of leaching, and radium is enriched here relative to uranium because it is less soluble. Further evidence that uranium is more readily leached than radium is presented in Chapter 6. It is also possible that radium is concentrated preferentially to uranium in plants which decay to form the A horizon, but there is no direct evidence to support this suggestion.

SAMPLING ERROR

DRAINAGE SURVEY

Two types of sampling error must be considered in drainage surveys. Sampling error in space is the variation in composition within a limited area near the sample point. Sampling error in time is the variation in composition at the same point over a period of time.

Sampling error in space was determined by collecting replicate samples, about 10 feet apart, at each of a series of 18 sample points.

Two water samples were collected at each point, two organic sediments at

10 points and two clastic sediments at 13 points. Each of the samples was analyzed in the normal way. Results are presented in Table 19. Standard deviation was calculated according to Appendix III. In order to achieve a random sample representative of the range of natural values the entries in the top three rows of Table 19 were omitted from the calculation.

Sampling error in time was determined by collecting replicate samples at different times, at least a month apart, from 24 sample points. Two water samples were collected at 23 points, and two clastic sediment samples at six points. No organic replicates were collected. Each of the samples was analyzed in the normal way and results are presented in Table 20.

A further estimate of the sampling error in time for radon in water was made by sampling two points a large number of times, although this method is felt to be inferior to collecting replicate samples at a large number of locations as described above. The writer collected 32 samples from a stream (31F4 8169) and 42 from Bow Lake. The first 19 lake samples were from sample location 31F4 5429 and the remainder nearby at sample location 31F4 3419. The radon level was essentially the same at the two Bow Lake sites. Results were reported by Smith and Dyck (1969). The standard deviation of radon levels in the stream is 15% and that for the lake is 30%. Means at both locations are about 120 pc/l, much higher than average. The sampling error in time of radon in water reported in Table 20, 57%, is more representative of the natural situation because it was based on a large number of different sample points, selected over the full range of radon concentrations.

Sampling errors are compared with analytical errors in Table 18,

15 H 13

Sampling Error in Space

Samples in top three rows were omitted from the calculations of standard deviations.

	10	Uranium pom		r~-	38		i. W		H	ကို	ių T	4.0	. 18	r1 -1	⊗	. 18			90.	8.	30 30 30
	ediments	Urani.		ង	14		ထ္ ဆု		.78	05,	æ; ∞	2.6	4	C)	ю Н	Parine Marine			29	ਨ <u>ਂ</u>	m
	Clastic Sediments	mg/od		9	∞ ~		₩.		ιĊ	-	라 :	, y	2	ë.	. 문	.12			27	. 12	18-20
	ğ	Radium pc/gm		r4 r4	근		83		D.1	拉拉。	ů.	φ Γ.	ij	97.	100	台			0.	8.	M M M
	1 0	Uranium ppm	48			220		99	0				က က		со С	3.	ŵ	in H	리		<u></u>
	diments	Urani	9			130 130		ထု	8				\ \ \ \ \		나- 살기,	<u>~</u>	بر ب	ത്	.50		<u> स</u>
	Organic Sediments	pc/gm	97			80		ं य	ri Ni				E.		o,	8.	о Н	æ.	æ.		
	රියි	Padium pc/gm	50			55		∞ ≈ i	2.7				a, a,		N.	ů	rad	, 0,	ď.		<u>2</u>
		quid turn	લ જ	37	디	က္	. 7	رب ش	7.2	Ä,	₹.	φ	ν.	۲-	Ġ	က္	ښ	ecopts	r-† •	O	00 E
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2 a ²	ig.	1 DC/7	1720		580	150 150	340	170	<u></u>	64	65	rd ar	ထု	8	57		2	Comme	ţ	0	No
	- 19 - 2]	Radon	1750	1720	650 00	G K	86 86	00 70 70	82	ο, Ω	% 0/	ထ္	2	9	1-4	()	파	-	Ó	O	n 14%
	scation er		T007	10007	4003	8055	8037	8069	80 07 07 13	8058	8067	TITO TO	8139	17.08 8057	8137 27	8135	00 00 11	87 10 10 10 10 10 10 10 10 10 10 10 10 10	9030	8120	Standard deviation
	Sample Location Number			の 日日 日日	(A)	31E1	31013	7年6		31016	記して	31HT	W. 1	E CO	TATE	3127	31	THE W	THO		Standard

TARIE 20

Sampling Error in Time

Samples in column 1 were collected between June 1 and August 1, 1968; column 2, between August 1 and September 15, 1968; and column 3, between June 1 and July 15, 1969.

				**						
Sample		Wa	ter	. At te	priz		Clas	itic S	ediments	
Location Number	Radon, 2	pc/l 3			ppio 3	Radi	ium, p 2		Uranium, 1 2	3 Dpm
31EL 8169	1 1 + 0		.,		~			2.4		8.2
31El 4001	12,100	1700		40	3.2					
31013 11162 260	300									
31F4 11667 70	43		* 3-	0 .						
3114 11792 66	42		1.6	. i.						
31016 8165	ZÚ	67	•	.9	.9		The state of the s	, 42	4.4	7.8
3184 11769 40	25		0	.2						
31116 8155	28	111		Annual second	.6				3.2	1.3
31E1 11163 23	29		Ò	1.2						
31174 20	29		.2	. 2						
3161 8100 33	3		0	. 3						
31174 8032 17		17	0		. 3					
3174 8181	19	16		0	. 7		.23	. 38	6.6	3.0
31P4 11789 8	17		0	0						
311/4 (013)	0	15		0	• 3		.10	.11	.18	. 16
3114 11714 10	16		2	P.						
3174 8082 13		Party Control	1.5		Lands,					
3174 8180	12	T\$		· war	0		.20	.20	1.8	.68
311786 8040 12		6	in the second		Ô					
31F4 8027 4		11	0		0					
31F4 11766 11			. ž	0						
31P4 11826 5	\$.									
31E1 11159 6	*		200 A							
31013 11157 ?	Ž,		I.	arus.						
Standard	in and the			33 <i>30</i>			ે <i>પૈર્મ</i>		<i>ા પ</i> ્રા	

117%

57%

deviation

36%

50%

Because each replicate was analyzed only once, the value presented for sampling error in space must be considered to be the sum of analytical error and sampling error in space. A significant sampling error in space will be evinced by a standard deviation greater than that for analytical error. Similarly, because sample points were not recorded to an accuracy of better than ten feet, sampling error in time must be considered to be the sum of analytical error, sampling error in space and sampling error in time. A significant sampling error in time will be evinced by a standard deviation greater than that for sampling error in space. Standard deviations are compared by means of the F distribution (for instance, see Freund, 1967, pp. 268-270).

Sampling error for radium in sediments is not significant because the standard deviation for sampling error is not much greater than that for analytical error. The fact that one of the values, 21%, is lower than the analytical error is evidence of the imprecision in determining these errors.

Sampling error for uranium in sediments is significant because all three sampling standard deviations are greater than analytical error. The difference in sampling error between uranium and radium is not significant except perhaps for organic sediments: the null hypothesis that sampling errors of 21% and 48% are equal can be rejected at the 90% but not at the 98% confidence level.

Sampling error in space for radon in water is not significant, but sampling error in time is. Sampling error in space is significantly lower for radon in water than for uranium in water, or for uranium or radium in sediments. This is probably due to the lower analytical error for radon. Sampling error in time is not significantly greater than

that for sediments. The advantage of the low sampling error in space is lost when one considers the sampling error in time.

The large sampling error in space for uranium in water is largely due to analytical error. It is significantly larger than the other sampling errors in space. The large sampling error in time for uranium in water is significantly larger than sampling error in space and the other sampling errors in time.

In Table 20 individual samples are divided into three groups corresponding to three different seasons. It is evident that no correlation exists between any of the elements determined and the season in which the sample was collected. This conclusion is substantiated, with respect to radon, in Figure 7.

Except for uranium in water, sampling and analytical errors are small compared to the range of concentrations found in nature. In Chapter 8, it is shown that geochemical surveys of uranium in water are useful in spite of the high sampling error.

SOLL SURVEY

At three locations in the soil survey duplicate samples were collected about ten feet apart. Each sample was analyzed in the normal way, and results are presented in Table 21. A surprising result is that the sampling error for radium in the A horizon is much lower than those for radium in the B horizon and uranium in the A horizon. Both these differences are significant at the 98% confidence level even with only three degrees of freedom. Although the situation is apparently reversed

TABLE 21
Sampling Errors in Soils

Sample Number	Urani		Radi	.m		
20 (% xxxxx) 41 (47 xxx)	A horizon	B horizon	A hor	rizon	B hor	rizon
31D16 7087	122 12	60 54	16	13	30	13
31D16 7085	13 8.2	40 - 44	16	19	40	15
31016 7083	11 6.6	5.4 3.2	26	24	15	4.0
Standard deviatio	n 73%	27%		L2%	(59%

for uranium, the A horizon giving a higher error than the B, this difference is not significant even at the 90% confidence level.

Sampling errors for soils are compared with analytical errors in Table 18. Because soils, on the average, are 10 times as high in radium as are sediments, the analytical error for radium presented in Table 18 is determined by considering only the top 16 pairs in Table 15, that is, the 16 pairs with highest radium content. Sampling errors for radium in the B and uranium in the A horizons are different from the respective analytical errors at the 98% confidence levels. Sampling errors for radium in the A horizon and uranium in the B are not, even at the 90% level. Compared to the range of concentrations found in nature, sampling errors in soil are low.

CHAPTER 6 SOME OBSERVATIONS ON THE GEOCHEMISTRY OF RADIUM AND URANIUM IN THE SURFICIAL ENVIRONMENT

Radium-226 in the lithosphere is closely associated with its parent uranium-238. Equilibrium in the uranium-238 decay series is essentially complete in a million years (Table 4), and rocks at Bancroft are much older than this. Rock containing 1 pc/gm radium-226 at equilibrium contains 2.9 ppm uranium-238 or, because uranium is 99% uranium-238, 2.9 ppm uranium.

The surficial environment in the Bancroft area has developed in the 12,000 years which have elapsed since the retreat of the Meistocene ice sheet (see Chapter 2). In 12,000 years radium will have reached something less than 0.002 of its equilibrium level with uranium (see Table 4). In other words, if a sediment were deposited at the close of the last ice age with 100 ppm uranium and no radium-226, it will now contain something less than 0.07 pc/gm radium*. Closer approach to equilibrium than this means either that the material was equilibrating longer than 12,000 years or that radium and uranium have similar geochemical behavior.

The degree of equilibrium in sediment and soil cannot be determined accurately by reference to routine analyses because radium analyses were carried out on the -10 and uranium on the -80 mesh fractions. Results of dissolving experiments (Tables 13 and 14), on the other hand, give an accurate indication of the degree of equilibrium.

As described in Chapter 4, four organic sediment samples were

^{*}This is equal to $100 \times \frac{1}{2.93} \times 0.002$.

first ashed and then dissolved. Radium and uranium were determined on raw samples, ashed samples and dissolved samples (Table 13). Most of the uranium-radium ratios in Table 13 are above 2.9, the equilibrium ratio, and the average ratio of fused samples is 15. Even this small amount of radium must have been precipitated: it has not had time to grow in from decay of uranium. From Table 18, the average ratio in water of ppb uranium to pc/l radium is roughly 0.2, that is, the opposite to that in organic sediment. Uranium is depleted in water and enriched in organic sediment relative to radium by a factor of $\frac{15}{0.2}$ = 75. Clearly both elements are precipitated by organic matter but uranium much more strongly.

A clastic sediment sample, collected about a mile downstream from Greyhawk mine, was sieved into seven size fractions and dissolved as described in Chapter 4. Each fraction was analyzed for radium and all but one for uranium (Table 14). The ratio of ppm uranium to pc/gm radium is fairly constant and has an average of 0.35 compared with an equilibrium ratio of 2.9. Two explanations of the depletion of uranium relative to radium are possible. (1) About 90% of the uranium has been leached from the sediments. Clastic sediments originate in the lithosphere in which the uranium decay series is in equilibrium. has been precipitated on the sediment. The first case would indicate clastic transport of radium and the second, chemical transport. Both elements are enriched in fine fractions relative to coarse (Table 14). This indicates either: (1) radium and uranium are transported physically, concentrated in heavy minerals which are relatively fine grained, or (2) radium and uranium are transported chemically and adsorbed to the surface of grains. Fine grains have a higher ratio of surface to wass, and hence

| 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000

more radium and uranium would be adsorbed to them.

The question of physical or chemical transport is suggested as a topic for further research. First, a heavy liquid separation would determine if uranium and radium were in the heavy minerals. If they are, physical transport is indicated; otherwise, chemical transport is suggested. This can be checked, in the case of radium, by making an autoradiograph of a thin section of a grain mount. If radium is adsorbed to grain surfaces, then it should be so detected in the autoradiograph. Uranium gives rise to less radiation than radium, and probably would not be detected.

Weathered surfaces of four outcrops were sampled, and the samples analyzed for radium and uranium (Table 22), the purpose being to determine which element, radium or uranium, is more readily leached. The rocks were from areas mapped by Hewitt(1957) as syenite, metagabbro, granite and hornblende paragneiss respectively. Locations are given in Appendix VI. Each sample was separated into two portions, a and b, ground and then dissolved with hydrofluoric, nitric and perchloric acids.

In all cases, ratios of ppm uranium to pc/gm radium are lower than 2.93 which would be found in unweathered rocks at equilibrium. Clearly, uranium has been leached from these rocks more thoroughly than has radium. A minimum of two thirds of the original uranium has been leached away, and if some radium has been lost, then more than two thirds of the uranium has been leached.

The high loss of uranium relative to radium from weathered rocks has an important application to uranium prospecting: outcrop samples

TABLE 22

Radium and Uranium in Weathered Rocks

Map Unit Hewitt, 1957	Sample Number	Portion	Uranium ppm	Radium pc/gm	Uranium Radium
Syenite*	31E1 3203	á	126	132	.95
States State Control of the		р	186	115	1.6
Metagabbro	31F4 3813	a	<0.5	1.06	<.5
		b	0.6	1.26	•5
Granite	3114 3818	\boldsymbol{a}	0.8	0.84	1.
AND ME LEADER		b	1.0	1.04	1
Hormblende	31016 3217	a	<0.5	.90	<.6
paragneiss		ь	0.9	1.14	.8

*Sample collected from an outcrop of radioactive pegmatite near Bicroft mine.

in the Blind River area were confused by high radicactivity and low uranium assays (Lang et al, 1962, pp. 128-29). "Joubin theorized that because exposures...were strongly radioactive but samples showed little uranium or thorium, these elements might have been leached from the outcrops, leaving strongly radioactive daughter elements". Had the outcrop sample been analyzed for radium-226 rather than uranium, the confusion would not have arisen. Fadium analyses are just as easy and give a much more accurate indication of the original uranium content than do uranium analyses. This does not apply where the uranium deposits being sought are appreciably less than a million years old, and the uranium decay series has not yet reached equilibrium.

CHAPTER 7 PROCESSES AFFECTING THE LEVEL OF RADON IN SURFACE WATER

LOSS OF RADON FROM SURPACE WATER

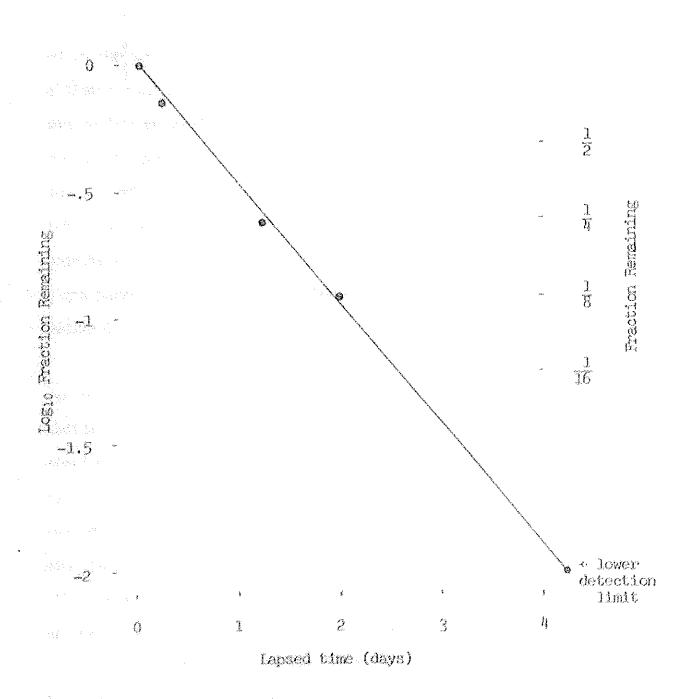
Two processes act to reduce the level of radon in surface water: radioactive decay and aeration. Radon-222, the chief isotope and the one considered here, decays radioactively with a half-life of 3.8 days.

Radon being a noble gas escapes rapidly from water when exposed to air. To determine the magnitude of this effect, an experiment was run to determine the residence time of radon in still water. A three gallon bucket with no cover was filled with water containing 97 pc/l radon and no radium, and left sitting outside. Radon in the water was measured four times in the next four days (see Figure 6). The last reading was below the detection limit of 1 pc/l. Plotting the logarithm of the fraction of radon left against the lapsed time results approximately in a straight line, that is, the decay is approximately exponential. The half-life of 16 hours is the time at which the fraction remaining is one half. As the half-life for aeration plus radioactive decay (16 hours) is much shorter than the half-life for radioactive decay alone (3.8 days), it is clear that aeration is more important in removing radon from water than is radioactive decay.

The bucket experiment represents a special case, and different values of half-life would be expected in nature. At depth, aeration would be less effective, and the half-life would approach that for radicactive decay, 3.8 days. In turbulent water, aeration would be more effective and the half-life shorter. The half-life of 16 hours would represent a maximum for shallow turbulent water and a minimum for still

FIGURE 6

Loss of Radon from Standing Water Exposed to the Atmosphere Initial concentration was 97 pc/l.



water at depth.

POSSIBLE SOURCES OF RAIXON IN SURFACE WATER

The short residence time of radon-222 in surface water, considered in the preceding section, indicates a constant and local source. The ultimate source is uranium in the lithosphere; the immediate effective source is radium-226. The question of the source of radon in water then reduces to the question of the location of the radium from which the radon is derived, that is, the location of the transformation of radium into radon and the route by which the radon arrives in the water. Four processes are suggested: (1) radium in solution, (2) radon travelling in a gas phase, (3) radon in groundwater influx and (4) radium in surface sediments.

The possibility of radium in solution accounting for the radon can be tested, in the Bancroft area, by reference to the literature. Radium in water has been measured by the Ontario Water Resources Commission (1968). A rough comparison of their results with those obtained by Dyck and Smith (1968) for radon shows that radon is present at a level 20 to 50 times that which would be expected in equilibrium with radium in solution. This result is confirmed in this thesis (see Table 18). Thus radium in solution does not account for a significant portion of the radon in water.

The possibility of radon travelling as a gas and then entering into solution in surface water is unlikely. Below the water table a mobile gas phase is most unusual, especially in Precambrian terranes, and

radon will not form its own gas phase (see Chapter 1). For radon to travel as a gas into a surface water body, the radium source must be at a hydrostatic level equal to or above that of the surface water.

That springs are frequently high in radon is well known and is substantiated by this work (see Figure 3 and Table 18). Undoubtedly influx of groundwater contributes radon to surface water. The important question is how far has this radon travelled, that is, how closely related is it to uranium leposits? The 3.8 day half-life of radon and the normal slowness of groundwater movement suggest a nearby source, within a few hundred feet. As this distance shortens to a few feet, addition of radon by influx groundwater grades into the fourth process, production of radon by decay of radium in sediments.

The contribution of radium in sediments to radon in water is evaluated in this thesis and found to be important. Radon emanation is the basis of the analytical method for radium. From Table 18 the median value for radium in sediments (i. e., radon emanation) is 0.6 pc/gm. The median value for radon in stream water is 23 pc/l. The amount of sediment required to supply this amount of radon to the water is calculated as follows.

Equilibrium in the pair radium-radon is approached with the halflife of radon (see Appendix I). In the case of radon in water exposed to the atmosphere, the effective half-life, allowing for aeration and decay, of 16 hours established above is used as a better approximation. Thus, after several days,

$$\lambda N_1 = \lambda N_2 , \qquad \dots (1)$$

where λ_1 is the decay constant for radium, N_1 is the number of atoms of radium, λ_2 is the decay constant for radon using the 16 hour half-life, that is,

$$\lambda_2 = \frac{0.6931}{16 \times 3600} \text{ sec}^{-1}$$
,

and N_2 is the number of atoms of radon.

In one liter of average stream water, there are 23 picocuries (pc) of radon. Because the picocurie is defined as the amount of radio-active substance in which 0.037 disintegrations occur per second, the number of atoms of radon present can be calculated according to

$$\lambda_3 N_2 = 23 \times .037$$
,

where λ_3 is the radioactive decay constant for radon,

$$\frac{0.6931}{3.8 \times 24 \times 3600}$$
 sec⁻¹.

Transposing,

$$n_2 = \frac{23 \times .037}{33}$$
.

Substituting this into Equation (1) gives

$$\lambda_2 \times 23 \times .037$$
 $\lambda_3 N_3 = \lambda_3$

the amount of radium required to supply 23 picocuries of radon. To change the units from disintegrations per second to picocuries, divide both sides by .037. This gives the amount of radium (in picocuries),

$$\frac{23\lambda_2}{\lambda_3} = 130.$$

Thus 130 pc of radium will supply the radon in a liter of non-turbulent "average" stream water. "Average" sediment contains 0.6 pc of radium per gram. Therefore, 220 grams or, assuming a density of 1.3, 170 cc of sediment will supply the above radon. The average stream sampled was 16 cm deep. A liter of water 16 cm deep has a base area of 62 cm². The necessary radium is available in the top 2.7 cm. This is about what would be expected from experience in the laboratory. In the analytical method for radium, radon is removed from a layer of sediment about 2 cm thick with a small amount of shaking. Although emanation is probably not 100% in the top 2.7 cm, some radon would come from deeper levels. Clearly there is enough radium in the sediments, on the average, to account for all the radon in the average stream water.

A theoretical model of the processes affecting the level of radon in surface water can now be proposed. Two effective sources of radon in surface water are indicated: radium in sediments and radon in groundwater influx. Two processes contribute to the loss of radon: decay and aeration. The remainder of the chapter is devoted to examining statistical relationships between the level of radon in water and several other variables. The fact that most of these relationships can be explained in terms of the above model reinforces its validity. Revelation of several unexpected relationships adds to the understanding

of the operation of the processes in nature.

EFFECT OF TEMPORAL VARIATIONS ON LEVEL OF RADON IN WATER

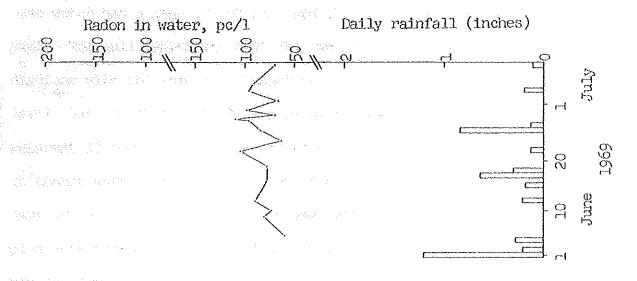
Ninety-one control water samples were collected to evaluate the effect of temporal variations in rainfall, wind velocity and flow volume on the level of radon in surface water. Most of these were collected in connection with the reconnaissance hydrogeochemical survey directed by A. Y. Smith mentioned in Chapter 1. Some of the results were presented by Smith and Dyck (1969).

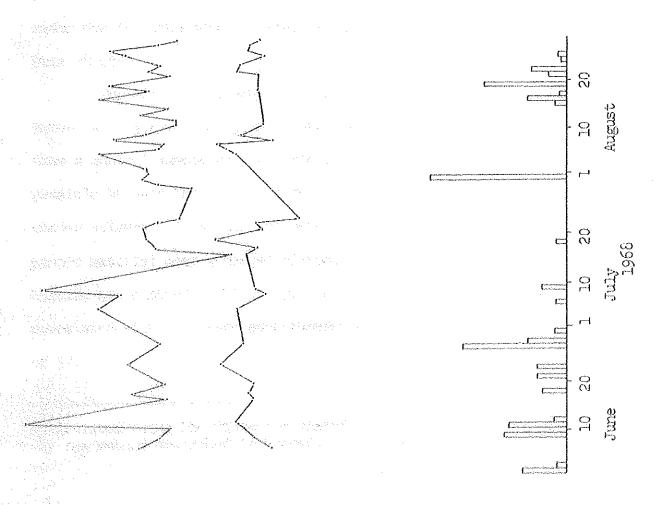
Stream sample point 31F4 8169 (see sample location map in appendix) was sampled by the writer 32 times in 1968. A point 200 feet downstream was sampled 17 times in 1969. Bow Lake water was sampled by the writer 42 times in 1968. The first 19 of these were from sample location 31F4 3429, and the remainder nearby at sample location 31F4 3419.

To evaluate the effect of rainfall on the level of radon in surface water, rainfall is plotted in Figure 7 against radon in water at the two control sample points. Rainfall was measured daily at Bancroft by the Canada Department of Transport. Two conclusions can be drawn from Figure 7. (1) There is no apparent correlation between daily rainfall and radon level. The drop in radon content of the stream water on June 24, 1969, following the heaviest precipitation day of the sampling period is probably fortuitous. Chamberlain (1964) reports a similar conclusion for uranium in surface water. (2) There is no apparent correlation in radon levels between the two control samples.

Because wind causes turbulence on open bodies of water, it was

Effect of Rainfall on Level of Radon in Surface Water





felt that radon level might be correlated with wind velocity as manifested by the presence of waves. In Figure 8 and Table 23, Bow Lake control samples are divided into two groups. The group of samples from turbulent water has a mean of 108 pc/l and that from non-turbulent water 126 pc/l. The null hypothesis that the two groups of samples came from populations with the same mean cannot be rejected at the 90% confidence level, but can at the 80%.* A more significant correlation might become apparent if more samples were collected. On the basis of this experiment it cannot safely be concluded that wind velocity, and hence turbulence, have any effect on radon level in lake water. Because the Bow Lake samples were from water about a foot deep, the weakness of the correlation may be explained by the waves stirring up sediments and releasing more radon to the water. This addition would tend to cancel out the loss of radon due to turbulence. A similar conclusion is arrived at later in this chapter.

The effect of change in flow volume on radon content of stream water, at a single point, was tested in the summer of 1969 when, each time a control sample was collected, flow rate was measured. This was possible because the control sample point was at the outlet of a square wooden culvert. Flow rate was determined by timing the movement of suspended material over a marked distance and measuring the water depth. Results are plotted in Figure 9. Neither radon nor uranium is strongly correlated with flow rate even though the flow rate varied by a factor of 33.

^{*}Calculated using the Student-t distribution with 24 degrees of freedom by the method described in Freund, 1967, page 256.

FIGURE 8

Effect of Wind on Radon Level in Bow Lake

Number of Samples

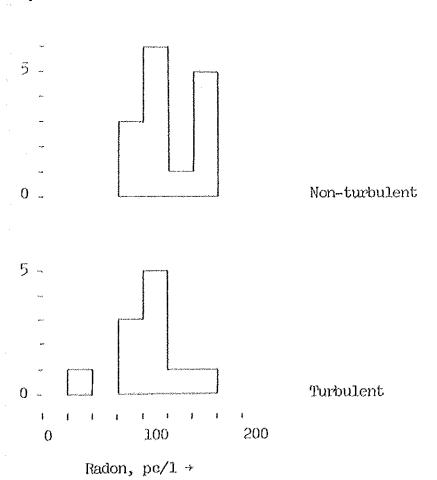


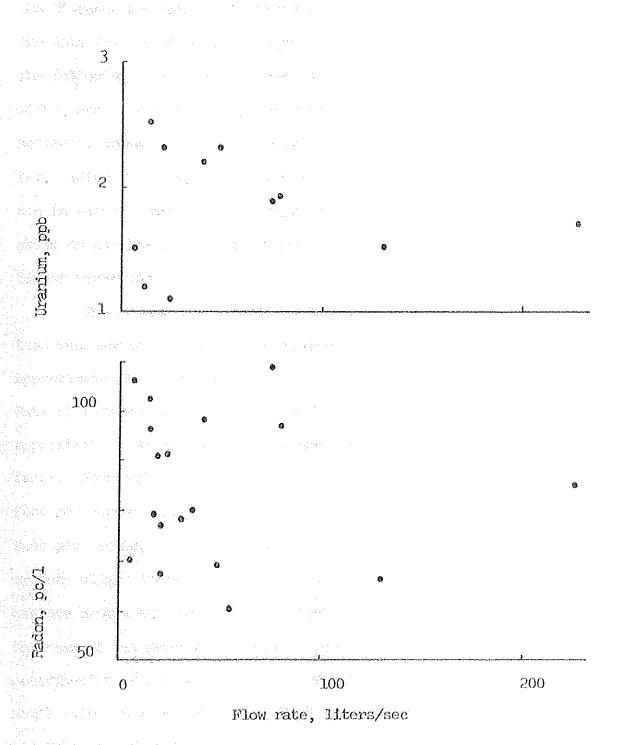
TABLE 23

Effect of Wind on Radon Content of Bow Lake

Sample Locati Number	on Date	Radon Con	Radon Content pc/1				
Muliver		Non-turbulent	Turbulent				
31F4 3429	July 5	167					
)I	16		37				
Į į	17	112					
n	19	122					
u	21		124				
1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	23	89					
en en skriver op de en	29		76				
31F4 3419	Aug. 2		123				
	5	168					
n n	6	108					
e de la companya de La companya de la co	7	105					
Ħ	8	155					
u.	9		123				
Alle Marie III	11	92					
	12		94				
u u	13	127					
ii ii	14		100				
i e e e e e e e e e e e e e e e e e e e	16	168					
a di M	18	116					
in the second	19	158					
	21		98				
	22	750					
	25		139				
1	56		159				
The state of the s	27		13.1				
orienis. La seconda	28	88					

RIGHER 9

Effect of Change in Flow Volume on Radon and Uranium Content of Stream Water, June and July, 1969



RELATIONSHIPS BETWEEN VARIABLES MEASURED FOR EACH SAMPLE POINT

As mentioned in Chapter 3, a standard Geological Survey of Canada 80-space geochemical field data card was filled out for each sample. The data are explained and listed in Appendix V. The correlation of the following variables with each other is tested in this section: width, depth, rate of flow, turbulence, flow volume, organic content of sediment, probability of contamination, temperature of water, pH of water, radium in sediment, uranium in sediment, uranium in water and radon in water. Then the individual and collective effect of these variables on the level of radon in water is tested by stepwise multiple linear regression.

The pH was measured with a Beckman model N pH meter. Temperature was measured in degrees centigrade. All the other observations are approximate visual estimates. Depth and width were estimated in feet. Rate of flow was determined at the fastest part of the stream by timing approximately the movement of suspended matter past an estimated distance. Four scale divisions were used: no visible motion, up to one foct per second, one to three feet per second and greater than three feet per second. Turbulence was noted using four scale divisions: no or very slight turbulence, ripples but water surface not broken, water surface broken and water falls. Organic content was estimated by eye to the nearest ten percent by volume. Contamination refers to potential contribution of the uranium series by cultural activities visible at the sample site, mostly road fill. Four scale divisions were used: none, possible, probable and definite. Flow volume was calculated by multiply-

ing width times depth times rate of flow.

SIMPLE CORRELATION COEFFICIENTS

The correlation coefficient r is calculated according to

$$n = \frac{n(\operatorname{Exy}) - (\operatorname{Ex})(\operatorname{Ey})}{\ln(\operatorname{Ex}^2) - (\operatorname{Ex})^2 \ln(\operatorname{Ey}^2) - (\operatorname{Ey})^2}$$

where n is the number of pairs of observations, and x and y are variables whose correlation is being tested.

A correlation coefficient of 1 indicates a perfect positive linear correlation between the two variables, that is, a line of the type y = ax + b, with a positive, will pass through all the points. A correlation coefficient of -1 indicates a perfect negative linear correlation, that is, as above with a negative. A correlation coefficient of 0 indicates that the variables are in no way correlated. Correlation coefficients between 0 and 1 indicate imperfect positive correlation; those between 0 and -1, imperfect negative correlation. Coefficients close to 1 or -1 indicate better correlation than those close to 0.

Simple correlation coefficients of the variables listed above are presented in a correlation matrix in Table 24. Data for 182 streams were treated separately from those for 49 lakes and swamps. The 11 samples from Paudash Lake are omitted because this lake is much larger than the others and may be contaminated. Where two sediment samples were collected, radium and uranium values used in Table 24 were determined by

Correlation Matrix for Variables Measured for Each Sample Point

Correlation coefficients between -.15 and +.15 (streams) and between -.28 and +.28 (lakes and swamps) can be attributed to chance at the 95% confidence level. Coefficients significant at the 95% confidence level. Coefficients significant at the 95% confidence level.

		width	depth	rate of flow	turbulence	flow volume	organic content of sediment	possibility of contamination	temperature (water)	pH (water)	In radium in sediment	In uranium in sediment	In uranium in water	In radon in water
		H	N	m		Ю	Ø	Ĺ	∞	Øλ	A	Γ-{ F-{	검	ద
182 Stream samples	5 6 7 8 9 10 11 12 13	.8212 .03 .11 .1013 - <u>.22</u> 06 - <u>.38</u>	.30 .01 .00 .21 .0902020117	51 30 11 05 20 11 15 08 29	90. 01. IO. 90. 80. IO. 70. IE 40.	17 .06 .06 .1112230143	.020302 .28 .28 .01 .06	<u>61. 81. 41. 81. 80</u> 90	32 .02 .3140303243	5409 .1915182116	. 46 . 22 - 39 - 50	.21 .122514 .43 .54 .64	.28045333 .44 .23	.26 .085735 .48 .18 .64
	∞ ⊐	.3307	106 - 24	와.			\ 	7	<u>ო</u>	0,	-,08	5	7	9
	N	.26	ľ		였.		%	7-	in the second	02	01.	8	- 35	in i
	Lake and swamples													

weighting each sample value by the relative amount of that material (clastic or organic) as estimated at the location. Natural logarithms were used for element concentrations because trace element concentrations are lognormally distributed (see Chapter 5 and Hawkes and Webb, 1962, pp. 25-26), hence using logs gives better correlation.

Correlation coefficients near zero may have arisen by chance. Significance of correlation coefficients is tested by the method described in Freund (1967, pp. 361 and 364). This method assumes that the variables are normally distributed random variables. Element concentrations are given normal distributions by using natural logs. It can be argued that some of the variables, for example, turbulence, rate of flow, possibility of contamination and flow volume, do not satisfy these assumptions. For correlations involving these variables, the following confidence limits are only approximate. The positive correlation between temperature and pH of water is probably due to the higher solubility of CO2 at lower temperatures. Dissolving CO2 in water lowers the pH. In the case of streams, the null hypothesis that any correlation coefficient between -.15 and +.15 is equal to zero cannot be rejected at the 95% confidence level. In the case of lakes and swamps, the range is -.28 to +.28. Correlations which are significant at the 95% confidence level are indicated in Table 24 by underlining.

A discussion follows of the reasons for the correlations shown in Table 24. Cause and effect relations among these variables are summarized in Figure 10 at the end of the chapter.

Reasons for the correlations in stream sample data between the following pairs of variables are obvious: flow volume with width, depth and rate; turbulence with depth (negative correlation) and rate; and

width with depth. For lake and swamp sample data, the correlation between depth and turbulence is positive. This is because shallow water near shore is protected from wind, and deep water near the center is open. In streams, shallow water causes turbulence due to interaction of the current with the bottom.

The negative correlations of organic content of sediment with rate of flow, turbulence and flow volume is due to the fact that low flow allows settling of fluffy organic material. The negative correlation of organic content of sediment with temperature, in the case of lakes and swamps suggests that the organisms responsible thrive in colder water. The negative correlation of pH with organic matter in these data is probably due to release by organisms of CO_2 or H_2S , or both (see Garrels and Christ, 1965, pp. 382-83). Reasons are unknown for the correlations of rate with width and pH, and of temperature with depth.

Correlations between radium and uranium are easily explained by a common source in the lithosphere. Also the correlation between radium and uranium in sediments is due in part to partial radioactive equilibrium in clastic sediments. Clastic sediments are chips of rocks, and rocks at Bancroft are in equilibrium (see Chapter 1). Therefore, except for differential leaching, clastic sediments are in equilibrium. Similarly, the correlation between radium in sediments and radon in water is due to local radioactive production of radon-222 from decay of radium-226 (see pages 88-90. The correlations of radon with uranium in sediment and water are due to the fact that uranium and radium have a common source in the lithosphere, and radon is produced from radium.

The affinity of uranium and radium for organic matter has been known for some time (see Chapter 1). The reason for the correlation

between uranium in lake and swamp water and organic content of sediment is unknown. The correlation of radon in lake and swamp water with organic matter in sediment is probably due to the concentration of radium in organic matter and the production of radon by radium. The negative correlations of radium and uranium in stream sediments with width, rate and flow volume are probably due to the fact that these variables are negatively correlated with organic content of sediment.

Radon and uranium in water, and radium and uranium in sediments are negatively correlated, in both sets of data, with water temperature. These negative correlations suggest that these elements are brought into the drainage by influx of groundwater. Groundwater maintains about the mean annual temperature and is colder, in the summer, than surface water. Dyck (personal communication) found a similar negative correlation between radon and temperature in the Athabasca, Saskatchewan, region.

The negative correlation between these elements and pH is unexplained. From the argument in the preceding paragraph, one would expect a positive correlation (see also page 110.

The reason for the negative correlation of uranium in lake and swamp water with depth is unknown. The positive correlation, in the stream data, of radium, radon and uranium with possibility of contamination indicates that radium and uranium are sometimes added to the drainage by cultural activity such as road building.

Radon is negatively correlated with depth, width, rate and flow volume. The negative correlation with depth is due to the fact that deep water is, on the average, farther than shallow water from the radon source in the sediments. In other words, the same emanation supplies radon to more water in deep than in shallow water. Depth of streams,

however, is highly erratic. Water depth at the sample point is not as important as average depth over a distance of several hundred feet upstream from the sample point. This quantity is difficult to measure. Flow volume is closely related to average depth and changes only at points of influx. It is easily determined by multiplying width times depth times rate, all of which are measured at the sample point. As one of these quantities changes, the others must change sympathetically to keep a constant flow volume. Measuring flow volume provides a more precise indication of average depth than does simply measuring depth at the sample point. Radon is more strongly correlated with flow volume (r = -.43) than with depth (r = -.17). A smaller random error is associated with measuring flow volume than with measuring depth, and random errors reduce correlation coefficients.

Width and rate are negatively correlated with radon in water because of their positive correlation with depth and flow volume. Correlation of radon with width (r = -.38) is stronger than with depth (r = -.17). This is because width, which is strongly correlated with average depth and flow volume, is less erratic than depth.

Because of the rapid loss of radon by aeration, it was expected that radon would show a negative correlation with turbulence. The lack of such a correlation probably indicates that turbulence stirs up the sediments, thus releasing radon and cancelling the effect of aeration.

A similar conclusion was reached on page 93.

Analysis of the data in this section was only partially successful. Some of the relationships are consistent with the model being tested (page 90), and some are not. Analysis of multivariate data by simple correlation coefficients suffers from the disadvantage that the variables are considered only two at a time. In stepwise multiple

linear regression, the technique used in the next section, the effect of many different variables on the level of radon in surface water can be considered at once.

MULTIPLE LINEAR REGRESSION

Multiple linear regression consists of fitting to the data an equation of the type

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \ldots + \beta_n X_n$$
,

where Y is the dependent variable (radon), X_n are the independent variables and β_n are constants. The degree to which the data fits the equation is measured by the multiple correlation coefficient which has the same qualities as the simple correlation coefficient discussed above.

The random error associated with a regression coefficient is expressed as a standard deviation. Dividing a regression coefficient by its standard deviation results in a t value. If the number of samples exceeds the number of variables by at least 50, the level of significance of a regression coefficient can be determined by entering its t value in the normal distribution. Thus the null hypothesis that a regression coefficient is equal to zero can be rejected at the 95% confidence level if its t value is greater than 1.96 or less than -1.96, and at the 99% confidence level if its t value is greater than 2.58 or less than -2.58.

Stepwise regression, the technique used here, fits the data to a linear equation one variable at a time. First, the dependent variable

and the most strongly correlated independent variable are fit to an equation of the type

$$Y = \beta_0 + \beta_1 X_1 .$$

The program² then selects the independent variable which, when added to the above equation, gives the greatest improvement in fit. The three variables are then fitted to an equation of the type

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 .$$

The program continues adding variables one at a time and calculating successively longer equations until there are no variables left which will give a significant improvement in fit. At each step it calculates, among other things, the multiple correlation coefficient, the constant term β_0 and, for each independent variable used, the regression coefficient and the standard deviation and t values of the regression coefficient.

Stepwise regression analysis was applied to the data from 182 stream samples using the 13 variables listed on page 97, with radon in water as the dependent variable. Again natural logarithms were used for

^{*}The program used is a subroutine (REGR) in Queen's WATFOR library, described in Queen's University Computing Center Technical Bulletin -6-L. It is a slight modification of the IBM S/360 General Program Library program number 3.4.003. In addition to the stepwise regression results described in this section, the program calculated the simple correlation coefficients discussed above. Computing was done at the WATFOR terminal of Queen's IBM 360 computer. L. H. Brockhoven of Queen's Computing Center advised on selecting the program, setting it up and interpreting the results.

element concentrations because they give a better fit. Results are presented in Table 25.

In step 1 the program selected radium in sediments as the most highly correlated independent variable. The strong positive correlation between radium and radon is due mainly to production of radon-222 by local decay of radium-226.

The best fitting equation using one independent variable is

In radon = 3.41 + 0.869 In radium.

because the regression coefficient is close to 1, the relationship between radon in water and radium in sediments is roughly linear. This supports the theory that much of the radon is derived from local radium. If all the radon were so derived and no random errors were involved, the regression coefficient would be 1.

Flow volume was added in step 2 as the variable which gives the greatest improvement in fit. The reason for the negative correlation was explained on page 103 Addition of this variable increases both the t value for radium in sediment (12.13 to 12.81) and the multiple correlation coefficient (0.671 to 0.758). This indicates that large flow volume reduces the radon level of water independently of its tendency to reduce the level of radium in sediment (Table 24).

In step 3 the program chose uranium in water as the independent variable which most improved the fit. From Table 4 it is evident that there is no cause and effect relationship between uranium and radon in water. The distributions of both uranium and radium are controlled by the distribution of uranium in the lithosphere and hence these two

Stepwise Regression Results for 13 Variables Measured for Each of 182 Stresm Sample Points

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	-		ដ	성용		etween -1.96 and 1.96 may have
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Step number	Multiple correlation coefficient	Constant	fileos monsent	Standard devlation of regression coefficien	value of egression	
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(i)	크용	8	(4) (1)	43 (D)	i ii	
iñ.	E 0	Ö	CC,	W FI	45.34	
			i saya -	494		
1	.671	3.41				
		1.0	. 869	.0717	12.13	In radium in sediment
2	.758	3.68				
F-W	ి. క్రాక్స్		. 815	.0636	12.81	In radium in sediment
	and the second second		025	.0034		
		10.78%	The Section of	• • • • • • • • • • • • • • • • • • • •	40.4.88000	
	.785	3.83				
- 5	. 100	3.03	.584	.0800	7.30	ln radium in sediment
						flew volume
			026	.0032	-7.96 4,41	in uranium in water
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11	.799	4.72			200	2 8 5 6 6 6 6 6 6 6 6 6 6 6 6 6 6 6 6 6 6
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			026	.0031	-8.13	flow volume
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5	.810	4,96		1.00	:	
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6 .819 4.69			
1. 18 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	illot.	4.07	ln radium in sediment
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022			flow volume
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2.054	.016	-3.37	temperature of water
092	.025	-3.66	organic content of sediment
1.23¶	.0870	2.69	In uranium in sediment
340			rate
.293	.1704	1.722	probability of contamination
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9 .826 4.39	a a ba	1. 60	2
. 1/26	4.8 とうこうしょう はんこう ひゅうかん	4,09 -6:24	In radium in sediment
022 022			flew volume In uranium in water
		2.95 -3.56	temperature of water
059 092		-3.68	organic content of sediment
.245		2,80	In uranium in sediment
370		1 8 40°C 193510 0	rate
261		The first of the control of the cont	probability of contamination
800.		$\tilde{1}.\tilde{1}\tilde{5}$	pH of water

variables are positively correlated. The distribution of radon is controlled by the distribution of radium in the sediments, and hence these two variables are positively correlated. The positive correlation between uranium in water and radon in water is thus indirect. This interpretation is supported by the fact that addition of the independent variable uranium in water to the equation reduces drastically the t value of radium in sediment (from 12.81 to 7.30). In this and all subsequent steps, the strong cause and effect relationship between radium in sediment and radon in water, so well illustrated in steps 1 and 2, is masked by the indirect relationship between radon and uranium in water, and the t value of radium is depressed.

Water temperature was the variable added in step 4. The reason for its negative correlation with radon in water was suggested on page 102. Apparently radon is added to the stream water by influx of cold groundwater.

Organic content of sediments was the variable added in step 5. The simple correlation coefficient of this variable with raden in water is insignificant, 0.06. The results of step 5, however, show that organic content of sediment has a significant negative effect on the raden level of water. Apparently organic sediment, being fine grained, or possibly due to adsorption, gives up its raden less easily than clastic sediment. (This effect is eliminated in the analytical method for radium by shaking the sample.) This negative effect was not evident from simple correlation coefficients because it was cancelled by the enrichment of radium in organic sediment. It was not previously suspected.

In step 6, the variable uranium in sediment was added to the regression equation. It is positively correlated with radon in water for the same indirect reason that uranium in water is, namely uranium

and radium in sediment are controlled by the distribution of uranium in the lithosphere, and radium gives rise to radon. The independent correlation of uranium and radium in sediment interferes further with the correlation of radius in sediment and radon in water and reduces further the t value for radium in sediment (7.11 to 4.07).

The program went through three more steps adding successively rate of flow, probability of contamination and pH of water. The effect of any of these variables on the regression equation may be attributed to random error. The correlation of pH of water with radon in water is positive in the regression equation and opposite to the simple correlation coefficient. A positive correlation can be explained as due to the addition of radon to the water by influx of groundwater. Groundwater in general has a slightly higher pH than does stream water (Garrels and Christ, 1965).

The program found that the variables turbulence, depth and width have a slight, if any, effect on radon in water and did not include them in the regression equation. Depth and width were left out because their contribution is included in flow volume. The lack of correlation of turbulence with radon in water is a surprise. The conclusion expressed on page 103 is borne out: turbulence increases the efficiency of transfer of radon from sediment to water, and this cancels its contribution to the loss of radon from water.

Several variables, notably uranium in water, have no direct cause and effect relationship with radon in water. Their correlation with radon interferes with the analysis of the effect of other variables on the level of radon in water. The stepwise regression program was run again for the same samples omitting the variables which could have no

effect on the level of radon in water and might interfere with the analysis. Uranium in water, uranium in sediment and rate of flow were omitted. Results are presented in Table 26.

The first two steps are, of course, the same as before. With uranium in water not available for the third step, the program selected organic content of sediment as the variable giving the greatest improvement in fit. Addition of this variable increases the t value of radium in sediment (12.81 to 13.86). That is, when organic content of sediment is considered, the relationship between radion in water and radium in sediment is atronger. This implies that both radium in sediment and organic content of sediment are controlling the same process which in turn controls the level of radion in water, namely the local production of radion in water by decay of radium in sediment. The effect of organic content on this process is negative, probably because organic matter traps some of the radio in the sediment.

Temperature of water is the variable added in step 4. Addition of this variable does not increase the t values of radius in sediment and organic content of sediment. This fact, together with the increase in multiple correlation coefficient (0.780 to 0.794), suggests that water temperature represents a separate process, namely addition of radon by influx of (cold) groundwater. The fact that the t value for radius is still substantial (11.64) proves that radon in water is strongly correlated with radius in sediment independently of their common negative correlation with temperature.

In the fifth and last step, the program selected probability of contemination as the only remaining variable which has a significant effect on the fit, and it could be rejected on the basis of its t value

TABLE 26

Stepwise Regression Results for 10 Variables Measured for Each of 182 Stream Sample Points

4 Table 1 Tabl				a - 1.
Step number Multiple correlation coefficient Constant term = 80	Regression coefficient = $\beta_{\tilde{c}}$	Standard deviation of regression coefficient	t value of regression coefficient	Natural log of radon in water is the dependent variable. Regression coefficients with t values between -1.96 and 1.96 may have arisen by chance at the 0.05 probability level.
1 .671 3.41	.869	.0717	12,13	In radium in sediment
2 .758 3.68	.815 025	.0636 .0034	12.81 -7.26	In radium in sediment flow volume
3 .780 4.12	.881 026 099	.0635 .0033 .0254	13.86 8.03 3.90	In radium in sediment flow volume organic content of sediment
4 .794 4.99	.789 026 091 056	.0678 .0032 .0249 .0169	11.64 -8.17 -3.67 -3.31	In radium in sediment flow volume organic content of sediment temperature of water
5 .800 4.59	.768 027 091 057 .369	.0679 .0032 .0246 .0167 .1761	11.32 -8.39 -3.68 -3.39 2.09	In radium in sediment flow volume organic content of sediment temperature of water probability of contamination

(2.69). The variables width, depth, turbulence and pilof water are found by the program to give a slight, if any, improvement in fit and are not included in the regression equation.

A hypothetical model, based on the physics and chemistry of redom (page 90), has been tested in nature by collecting relevant data. All the results of stepmise multiple linear regression analysis of the data are consistent with theory: this lends strong support to the theory. Spurious simple correlation coefficients are due to the shortcomings of simple correlation.

The processes affecting the level of radon in stress water can now be suggested. These processes are listed, in order of decreasing importance, in Table 27. Also listed, for each process, are the variables, measured for each semple point, which control or measure the process. Production of radon in water by decay of radium in sediment is controlled by the level of radium in sediment and by the organic content of sediment. Influx of radon in groundwater is measured by water temperature and possibly pli. Loss of radon by aeration and decay is controlled by flow values: the ratio of water values to bottom area (the source of radon) is higher in deep, large-flow streams than in shallow, small-flow streams.

case and effect relativaships among the variables measured for each sample point plus influx of groundwater and ununium content of the lithosphere are augmentized in Figure 10. Direction of the cause and effect process is indicated by an arrow. Uranium content of the lithosphere has a controlling effect on ununium and radium in the drainage. The half-life of radon is too short for it to nove directly, except under unusual directances as in some radon-rich springs and mine waters, from the lithosphere to the drainage.

TAGE 27

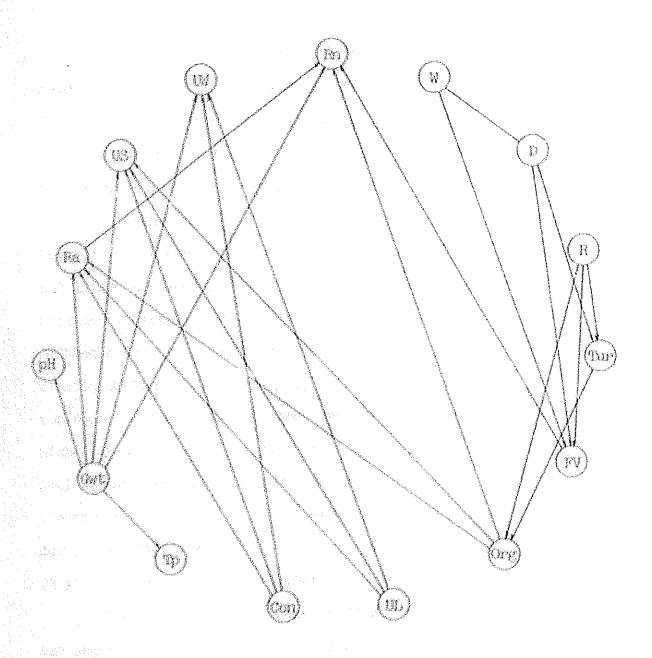
Summary of Processes Affecting Level of Radon in Streem Water

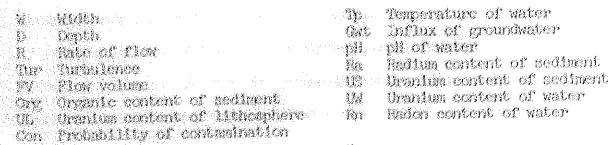
光海港 医电影 化自己分配 医二十二氏 医动物管 电影动力 医

		Index variable beasured for sample point
	production of radon in water by decay of radium in sediment	radium content of sediment organic content of sediment
(2.)	loss of radon by seration and decay	flow volume
	influx of redon-charged grandwater	water temperature pli of water?

MIGUTE 10

Cause and Rifect Relationships Related to Level of Redon in Stream Water





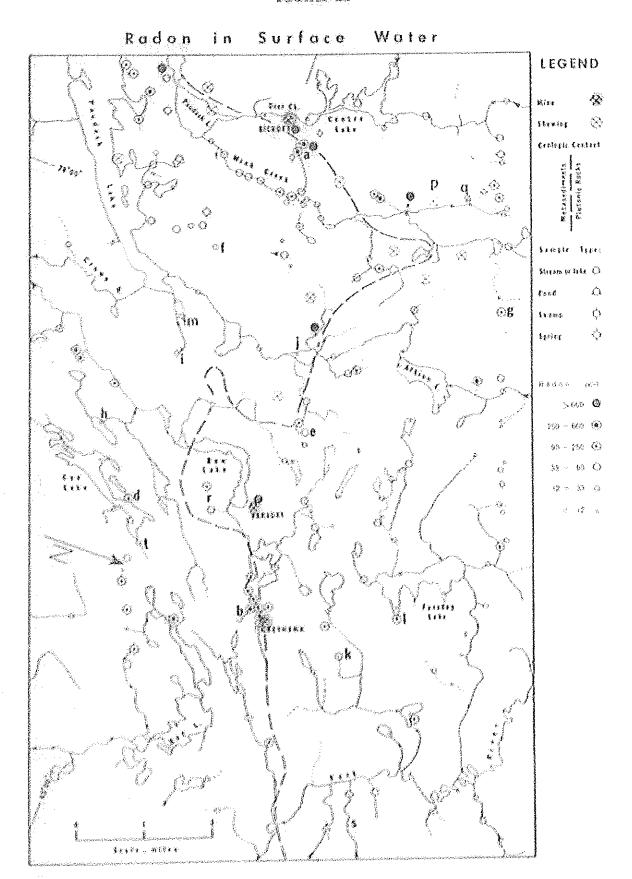
CHAPTER 8 INTERPRETATION OF DRAINAGE MAPS AND RECOMMENDATIONS FOR GEOCHEMICAL PROSPECTING

Four different uranium prespecting methods using drainage are suggested in Table 18: radium or uranium in sediments, or, radon or uranium in water. Radium in water is so low relative to the detection limit that its use as a prospecting tool is ruled out. This chapter compares the usefulness of these different methods.

Response of the different elements in water and sediment to imanium deposits and rock type is compared by plotting regional geology and analytical results on a series of base maps (Figures 11 to 16, also included in pocket). It is evident from Table 18 that data for organic sediments should not be plotted without qualification on the same map as clastic sediment, otherwise a high value due to the proximity of a uranium deposit could not be distinguished from one due to the relatively high uranium centent of organic sediments. For this reason it is necessary to make two maps each for uranium and radium in sediments: one for clastic and one for organic. A method of combining the two types of data, described later in the chapter, results in two more maps (Figures 19 and 20, also included in pocket).

The common base map shows drainage, simplified geology and uranium deposits. Different types of drainage sample points—streams, lakes,
smamps, ponds and springs—are distinguished on the maps by four types
of symbols as well as the chvicus difference in drainage pattern between
takes and streams. Points not on the drainage pattern and with no ticks
represent intermittent streams. Pends are defined as lakes which are
too small to show on the National Topographic Series, 1:50,000 maps.

FIGURE 11



Ú

FIGURE 12

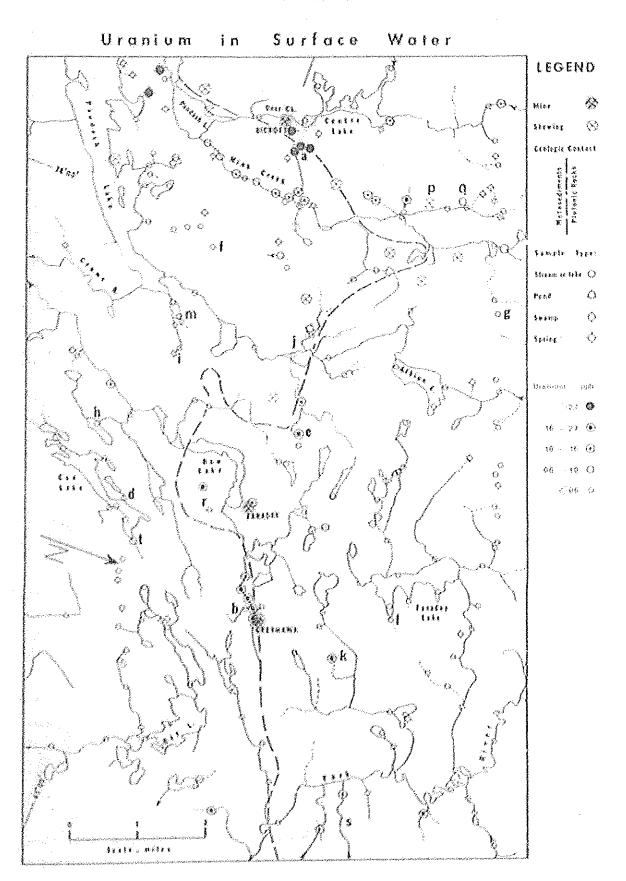


FIGURE 13

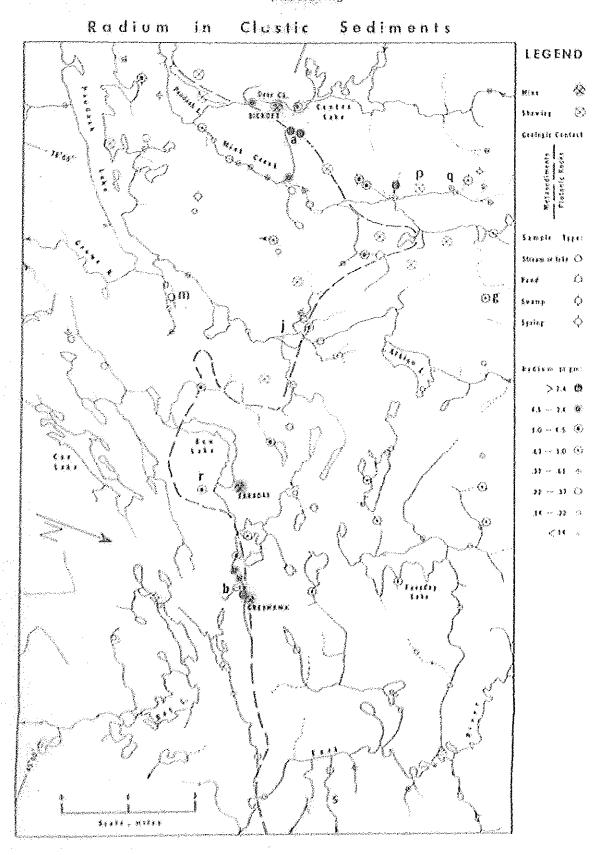


FIGURE 14

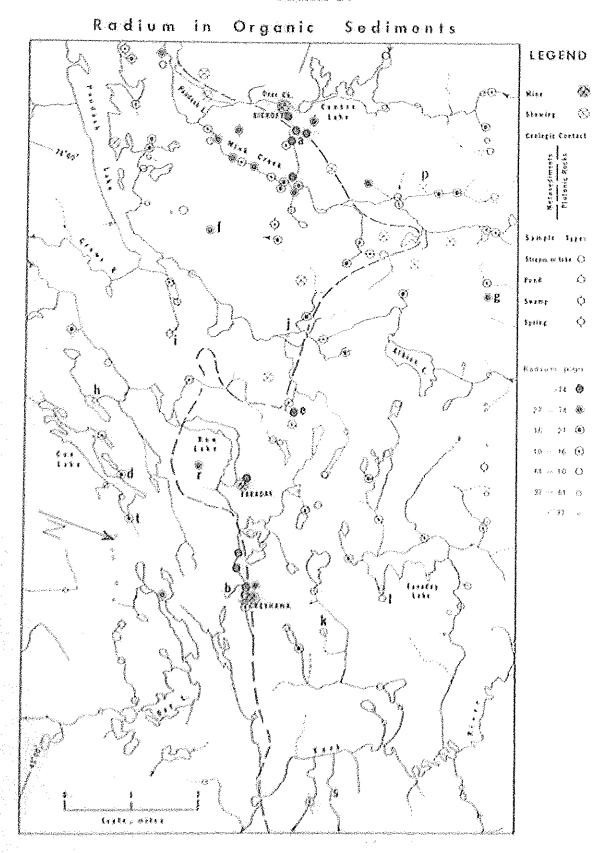


FIGURE 15

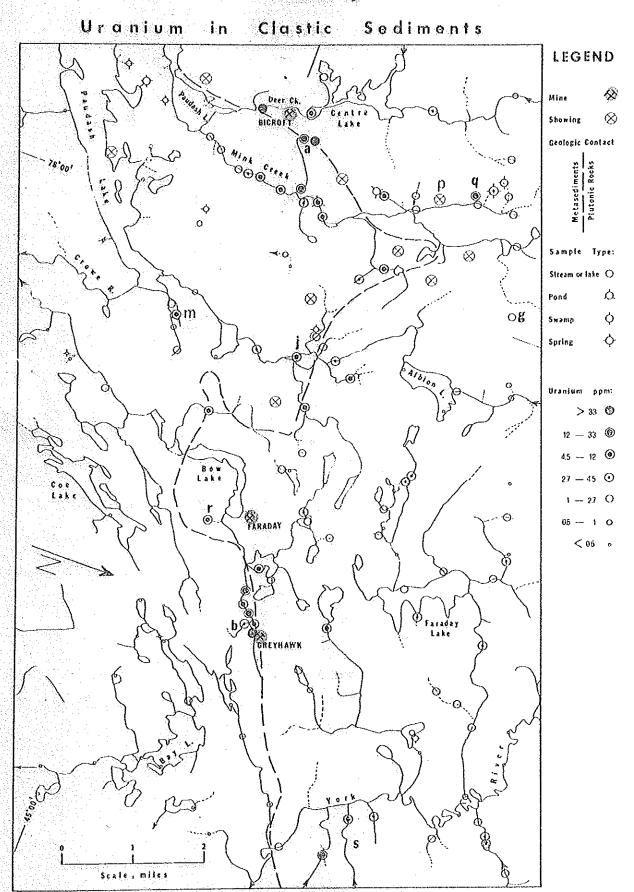
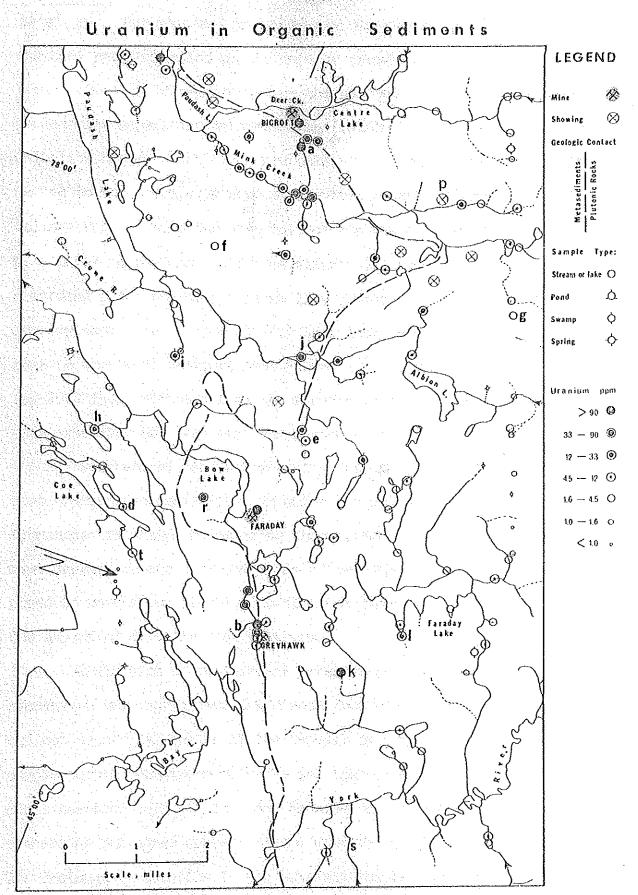


FIGURE 16



Where not obvious from the drainage pattern, flow direction of streams is given by arrowheads. In two places the course of streams is unknown, and they are terminated on the maps by arrowheads.

Geology is represented on the geochemical maps by a single heavy broken line separating metasediments (mainly marble, paragneiss and amphibolite to the south) from plutonic rocks (mainly granite, syenite and gabbro to the north). A zone of syenitic rocks and nepheline gneiss follows this line in the west half of the map and parallels it about a mile north in the east half (see Figure 1). Uranium deposits are concentrated along the heavy broken line. Major ore bodies are found in three places (see Chapter 2): (1) in a long zone extending from Bicroft mine to the Croft workings (see Figure 1), (2) in a limited area extending 2500 feet southwest from the Faraday shaft, and (3) in a limited area extending 500 feet east and northeast from the Greyhawk shaft.

Geochemical prospecting using drainage is based on the principle that element concentrations in water and sediment are higher near and downstream from mineral deposits then elsewhere. The degree of enrichment depends on the size and grade of the deposit, the amount of it exposed to weathering agents, its distance from the drainage channel and the amount of dilution from upstream.

Evaluation of geochemical prosperting techniques is based on the assumption that enrichment downstream from mineral deposits is natural and not caused by working of the deposit by man. Information for the sample points downstream from the tailings ponds at Bicroft and Faraday mines has been left off the maps because of the possibility that ore processing has added members of the uranium decay series to the drainage. The drainage in question is Bow Lake and Paudash Lake and the streams

draining them. Tailings from the Bicroft mill were piped across Deer Creek to a tailings pond near the headwaters of a small tributary. Any leakage from this tailings pond would have entered Deer Creek downstream from the sample point shown on the map. Contamination downstream from Greyhawk mine is unlikely because ore was milled elsewhere.

Scale and sampling density must be kept in mind when comparing prospecting techniques. Results of this survey, in which samples were collected at a density of about 2.5 per square mile over an area of 90 square miles, are not directly comparable with those reported by Smith and Dyck (1969), for which samples were collected at a density of 1 sample per 2 square miles over an area of 1650 square miles. Nor are the results of the drainage survey, presented in this chapter, directly comparable with those of the soil survey presented in Chapter 9. Prospecting methods recommended for geochemical surveys at different scales are summarized in Table 29 in Chapter 10.

The maps (Figures 11 to 16) show that all the methods studied are useful in prospecting for uranium. All the maps show a positive correlation of high values with the contact between plutonic rocks and metasediments, along which known uranium deposits are concentrated. All the maps show a positive response to Bicroft and Greyhawk mines. In the stream which rises one-half mile northeast of Bicroft mine (point a), water and both types of sediment are high in all the elements measured, in response to the northward extension of the Bicroft radioactive zone. The same is true for the stream which runs past Greyhawk mine downstream from the mine. Although it may be argued that mining activity at Greyhawk mine has altered the natural distribution of elements, this is considered unlikely for reasons given above. The chances of contamination

at point a are negligible because activity here was limited to trenching and diamond drilling and this had ceased ten or more years previous to this study.

As a geochemical prospecting medium, sediments are clearly more useful than water.

Levels of radon and uranium in lake water are correlated with neither proximity of uranium deposits nor rock type. Uranium is higher at the north end of Centre Lake, a mile north of Bicroft mine, than at the south end which is much closer. The opposite is true for sediments, in which both radium and uranium are much higher, near the mine than a mile north. Pond water, on the other hand, can be used in geochemical prospecting at this scale. Water in the pond near Bicroft mine is very high in both radon and uranium. Ponds are more useful because, being smaller, their water has a more local source.

The negative contribution of lake water as a geochemical prospecting medium at this scale does not contradict the conclusions of Smith and Dyck (1969). They found that levels of both radon and uranium in lake water were positively correlated with broad areas of uranium occurrences. The anomaly they showed associated with the Bancroft uranium camp is almost as large as the whole area studied in this thesis. Because lake water samples can be collected cheaply and rapidly using aircraft, they are useful in reconnaissance prospecting, but there is no point in collecting samples at the present density.

At the scale used in this thesis, stream water is more useful in prospecting for uranium than is lake water. Water in the stream which rises one-half mile northeast of Bicroft mine (point a) and water downstream from Greyhawk mine are high in both radon and uranium.

Stream sediments, however, are again more useful than stream water.

Water in Deer Creek downstream from Bicroft mine has background levels of radon and uranium; however, sediment at the same location is high in radium and uranium. Flow rate in Deer Creek is the highest in the area except for the York and Crowe rivers. Uranium-poor water from Centre Lake, a half mile upstream, dilutes any local influx of uranium-rich water. The low level of radon has two possible explanations: (1) radon-poor water travels the half mile from Centre Lake so fast that it doesn't have time to pick up a large amount of radon from the sediments of Deer Creek, which is swift and turbulent, or (2) the large flow volume indicates a high ratio of average depth to area of stream bed which results in a low ratio of radon in water to radium in sediments (Chapter

The small stream shown near Faraday mine rises in a spring directly above the workings. Its water is high in radon and its sediment is high in both radium and uranium. Uranium in the water, however, is lower than in several streams not associated with known uranium deposits. The reason for this is not known.

The stream which flows past Greyhawk mine has a tributary (point b) entering it from the south about a quarter of a mile downstream from the mine. Water in this tributary is high in radon but the sediment is low in radium. No uranium deposits are known in this area. Flow volume at this sample point is one of the lowest in the area. The stream is less than an inch deep, less than a foot wide and barely moving. The high level of radon in the water in spite of the low level of radium in sediment illustrates the concept developed in Chapter 7: the low flow volume and hence average depth result in a low ratio of water to emanations, thus a high ratio of radon in water to radium in sediment.

At three key places—Deer Creek, Faraday mine and the tributary downstream from Greyhawk mine—stream water sampling and analysis give misleading results. Stream sediments at these places give "correct" results. Similarly, sediment in Centre Lake gives a more accurate indication of the proximity of the Bicroft uranium deposit than water does. Thus, Walker's (1968) generalization on hydrogeochemistry (see Chapter 1) is true for uranium and radon in water: "...this technique is generally a cumbersome method of stream sediment sampling."

Sediments should generally be used in preference to water except for two cases: (1) in reconnaissance prospecting where lake water can be sampled cheaply and rapidly by aircraft, and (2) where rapid analytical feedback is possible and necessary. Measuring radon in water is the fastest

technique available—a portable radon—detecting apparatus is available* which permits on—the—spot analysis. Measuring radon in sediments might be just as rapid as and more effective than in water, but has not been tried. It would be less effective than measuring radium in sediments by the writer's method. The choice between measuring radon and uranium in reconnaissance lake water surveys should be based on logistics. If a small laboratory can be set up in the field, and if rapid analysis is important, then radon should be used. If samples must be sent to a central laboratory, then uranium must be used.

Two kinds of sediment, organic and clastic, are found in the Bancroft area (see Chapter 3). Both types were found at about one third of the sample points, clastic alone at one third, and organic alone at one third. Both radium and uranium are enriched in organic sediments relative to clastic (Table 18). Analytical results are plotted on four maps (Figures 13 to 16), two each for radium and uranium.

Examination of the maps shows that both radium and uranium respond to rock type and distribution of uranium deposits more accurately in clastic than in organic sediments.

For example, one would expect radium and uranium levels in Mink Creek to drop off gradually as it leaves the area of uranium deposits and flows towards Paudash Lake, that is, along the portion of the map near the label "Mink Creek". For both elements the drop off is more regular in clastic than in organic sediments. The trend reverses for radium at the farthest downstream sample point where clastic sediment is high.

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^{*}Bondar-Clegg & Company Limited, 768A Belfast Road (M.R. 1), Ottawa 8; McPhar Geophysics Limited, 139 Bond Avenue, Don Mills, Ontario; S.R.A.T., 41 rue Emerlau, Paris 15e, France.

This point is connected to the Bicroft mine area by a low swampy area through which radium may have moved into Mink Creek.

At several points not related to known mineralization, organic sediments are high in radium or uranium but not both. These points are indicated on the maps by the letters d to g for radium, and h to 1 for uranium. The anomalies are believed to be spurious, that is, not related to uranium in the lithosphere but due to local chemical conditions in the drainage. Support for this interpretation is given by the fact that these samples are high in only one of the two elements and low in the other. Similarly, clastic sediment at location g is fairly low in radium and at location j, fairly low in uranium. With the possible exception of points e, j and k, discussed later, further investigation of these spurious anomalies for economic reasons is not recommended.

The affinity of radium and uranium for organic material in sediments (Chapters 1 and 5) apparently is strong enough, in some cases, to mask the relationship between radium and uranium in organic sediments and in the lithosphere. This does not mean that organic sediments should be overlooked. A survey using only clastic sediments would leave unacceptable gaps in the coverage because they are not everywhere present.

Several anomalies in clastic sediment, not related to known uranium deposits, need to be explained. The high level of uranium in clastic sediment in the small lake (m) at the northeast end of Paudash Lake is probably due to contamination from road fill. Scattered high values along the line joining Bicroft mine with the Croft workings (p) are due to the northward extension, along this line, of the Bicroft

pegmatites (see Chapter 2). The high uranium value in clastic sediment nearby at point q may be explained in part by the fact that this sediment contains 30% organic material.

Five anomalies, still unexplained, are worthy of further consideration, but none of them are as intense as those associated with the mines. The most interesting is south and east of Bow Lake at point r, where two sample points have sediment high in radium and uranium, and one has water high in radon and uranium. The second is the triangle formed by Albion Lake and points e and j, which contains several high values. The third extends off the northeast edge of the maps at point s where three streams draining from the northeast into York River contain high values. The fourth is at point k north of Greyhawk mine, where water and organic sediment are high in uranium. The least interesting anomaly is at point t at the northeast end of Coe Lake, where organic sediment is moderately high in uranium and radium.

The lack of intense anomalies is no surprise. The area has been well prospected by both amateurs and professionals and has been mapped geologically at a scale of two inches to the mile. This was one of the reasons for selecting the area. Probably all the uranium deposits with good surface exposure have been discovered. Discovery of anomalies was not a primary purpose of this thesis.

Interpretation of sediment surveys can be facilitated by plotting the results for both organic and clastic sediments on the same map. Results for organic sediments cannot, however, simply be added to maps

showing radium or uranium in clastic sediments. Relationships between organic and clastic sediment at the same locations are shown in Figure 17 for radium and Figure 18 for uranium. Logarithmic (base 10) scales are used in both cases because they give a better fit. Best fitting straight lines were estimated visually in both cases. Equations of these lines, together with correlation coefficients (0.83 for radium and 0.74 for uranium), are shown on the figures.

The line which best fits the two types of radium data is

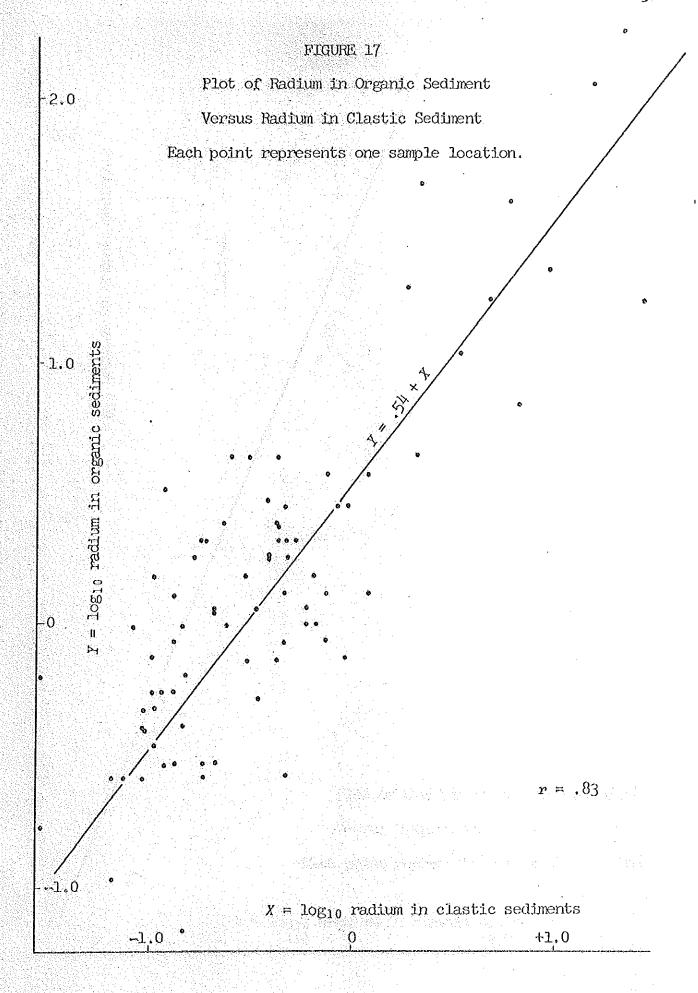
$$Y = 0.54 + X$$

where X and Y are logarithms (base 10) of the radium content, in picocuries per gram, of clastic and organic sediments respectively. Radium
in organic sediments is, on the average, 3.5 (the antilog of 0.54) times
as high as in clastic sediments at the same location. Thus, before
plotting the results for organic sediments on the same map as those for
clastic, it is necessary to reduce them to the same "scale" by dividing
the level of radium in organic sediments by 3.5. Figure 19 shows the
results for radium in the two types of sediment plotted on the same map.
For sample points with both types of sediment the results for clastic
only were used. Where organic sediments only were present, the radium
value was divided by 3.5 and the result was plotted.

A more complicated correction procedure was needed for uranium data. The best fitting line shown in Figure 18 is

$$Y = .18 + 1.27X$$

where X and Y are logarithms (base 10) of the uranium content, in parts



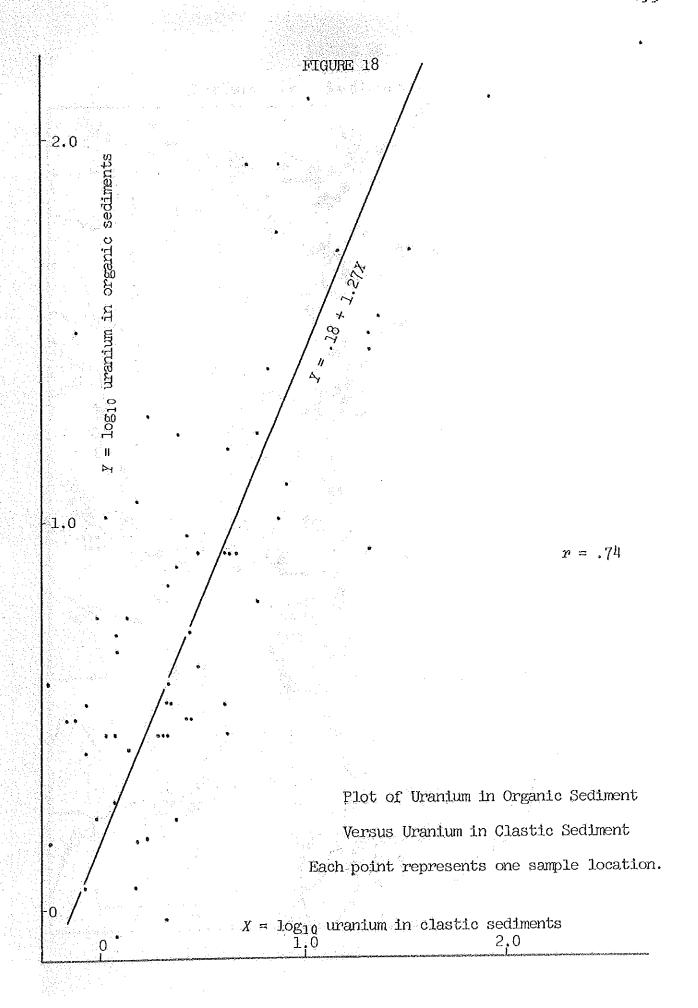
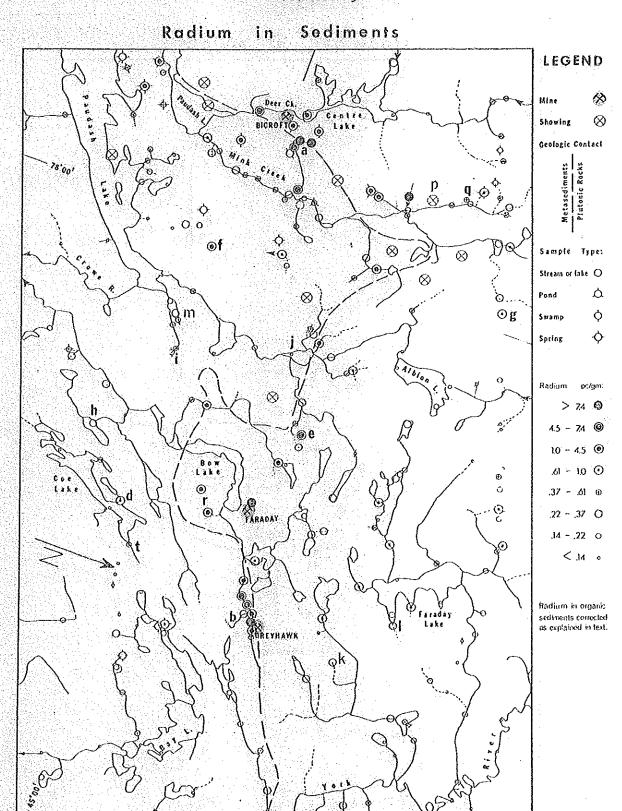
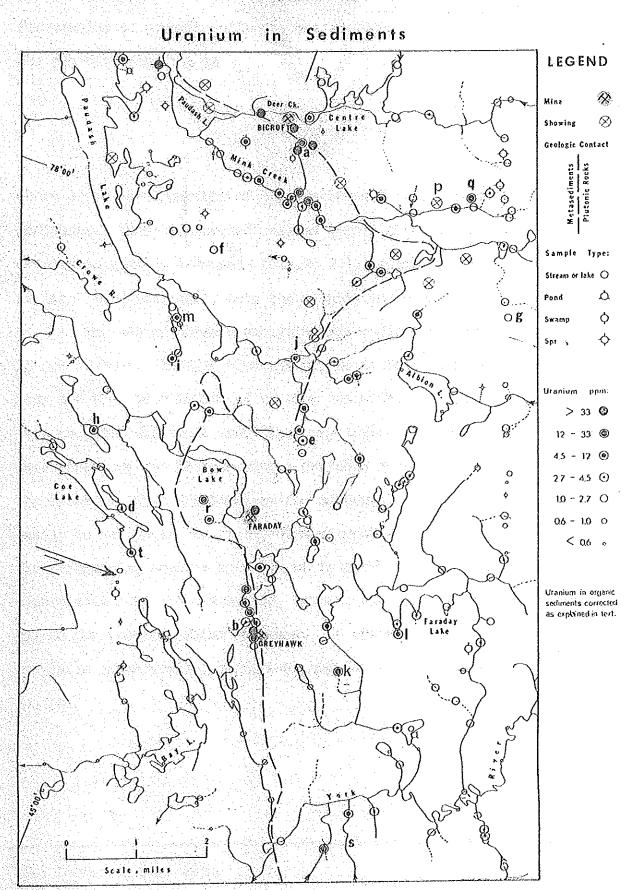


FIGURE 19



Scale, miles

FIGURE 20



per million, of clastic and organic sediments respectively. Results for uranium in organic sediments were reduced to the same scale as those for clastic according to

$$x' = \frac{\ddot{x} - .18}{1.27}$$

where X' is the logarithm of the "corrected" value of uranium in organic sediments. These "corrected" values were plotted on the same map as uranium in clastic sediments (Figure 20). Again, wherever both types of sediment were available, only the results for clastic were used.

The choice between analyzing for radium and uranium in sediments is not obvious. The two elements appear to respond equally well to geology and the distribution of uranium deposits. Results of larger surveys or under different conditions may indicate a preference. Radium determinations can be made with a portable radon apparatus which is on the n. ket.* Thus a laboratory for determining radium in sediment could easily be set up in the field, even without electricity. The apparatus for determining uranium in sediment is much less portable and requires electricity. On the other hand, several days equilibrating time is required for precise radium analyses. At present, radium analyses are not available commercially; therefore, the industry is generally limited to uranium.

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^{*}See footnote, page 128.

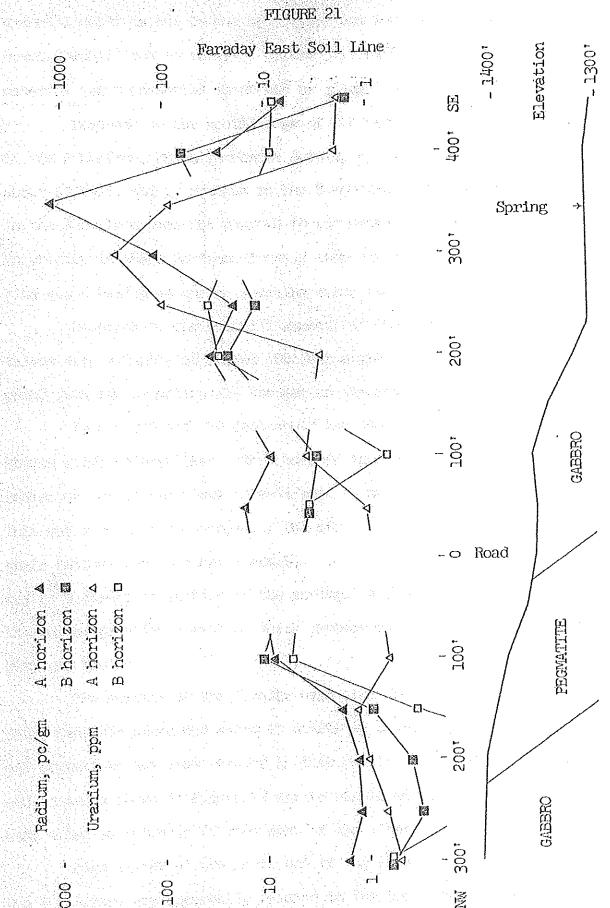
CHAPTER 9 INTERPRETATION OF RESULTS OF SOIL SURVEY AND RECOMMENDATIONS FOR GEOCHEMICAL PROSPECTING

Two approaches must be taken to the interpretation of the results of the soil study. First, applications to detailed prospecting are established by comparing the distribution of radium and uranium in soils to bedrock geology as known from mine workings. Second, applications to reconnaissance prospecting are established by comparing levels of radium and uranium in soils over mines with those in "background" soils collected some distance from known uranium deposits.

Distribution of radium and uranium in soils is compared to bedrock geology at the Faraday east line in Figure 21 and at the Faraday west line in Figure 22. The figures show, on a logarithmic scale, levels of radium and uranium in the A and B horizons at each sample point. Gaps indicate places where no soil was available. Lines intersecting the bottom of the figure indicate analyses below detection limits. Distribution of rock types and ore zones is based on conversations with and mine plans provided by R. Moss of Can-Fed Resources Corporation. Contacts were projected unwards from the adit level (elevation 1180 feet), using a strike of N 70° E and a dip of 55° S. Surface elevation and other features are also shown. Ore zones and pegmatites at Bicroft are too irregular to project to surface. Because the property was inactive, no mine staff were available for consultation.

The surface at the Faraday east line (Figure 21) slopes continuously to the south. Underlying bedrock is gabbro and one pegmatite dike.

A strong geochemical response to the uphill edge of the pegmatite is apparent in the soil, shifted about 100 feet downhill. The



downhill shift is due to one or both of two reasons: (1) material has moved downhill due to gravity, either physically or in solution, (2) material was transported southward by glacial activity.

Response to the uphill edge of the pegmatite is shown by radium in the A horizon, which increases 6-fold, by radium in the B horizon, about 40-fold, and by uranium in the B horizon, about 15-fold. Uranium in the A horizon does not respond to the pegmatite. The 2-fold increase in uranium in the A horizon directly over the uphill edge of the pegmatite could easily be due to sampling error (see Chapter 5).

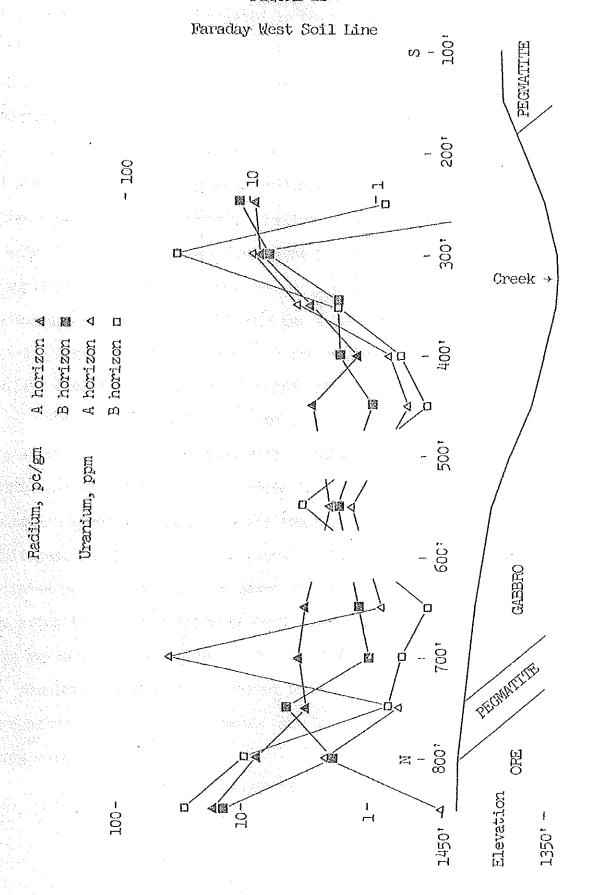
Response to the downhill contact of the pegmatite is marginal. Values drop slightly at 50 and 100 feet south. Apparently material has moved from the pegmatite all the way to the south end of the line.

Points 300 and 350 feet south are covered by a swamp, and a spring rises between them. No B horizon is present. Both uranium and radium values in the black organic soil at this point are extremely high. This may simply be an example of the affinity of both elements for organic matter (see Chapters 1 and 5). On the other hand, the high values may be a direct reflection of the spring. A diamond drill hole near the spring provides a path by which groundwater might be communicating with ore bodies.

The surface at the Faraday west line drops from north to south until near the south end where it starts up a small hill. Ore bodies and pegmatites are less regular in this part of the mine than elsewhere, and contacts shown in Figure 22 may be misplaced by 50 feet. The ore body shown is actually 50 feet east of the line.

High values at the north end of the line, except for uranium in the A horizon, are apparently related to the ore body. The uranium peak

FIGURE 22



in the A horizon at 700 feet is unexplained. High radium values south of the intermittent creek are related to the pegmatite uphill. The fact that uranium is lower at 250 feet than at 300 feet suggests that the uranium peak at 300 feet is related to the intermittent creek rather than to the pegmatite.

Some general conclusions are evident from Figures 21 and 22. Radium and uranium in the B horizon and radium in the A horizon are useful in detailed prospecting for uranium. Uranium in the A horizon bears no relation to bedrock and little to uranium in the B horizon or to radium. It has no application to detailed prospecting. In the B horizon, radium and uranium are equally useful. Should the A horizon be collected in a detailed survey, it should be analyzed for radium rather than uranium. A similar conclusion regarding weathered rock was reached in Chapter 6.

The usefulness of radium and uranium levels of soils in prospecting at a wider scale for uranium is established by comparison of these levels in soils over mines with those in "background" soils. Time limitation precluded collecting different type of soil over different rock types. An indication of regional background was obtained by sampling three points about 20 feet apart at the southeast tip of Monck Lake. This point is a mile from the nearest known uranium occurence. Nearby rocks are mapped by granitic by Hewitt (1957). Both A and B horizons were sampled. Results are expressed in Table 28 where each of the first three columns represents a separate sample. The range is an indication of regional background.

Support for this background is given by the three northernmost

TABLE 28

Background Values for Radium and Uranium in Soil ---Bancroft Area

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	and the organization of the		*	Range
Uranium ppm	A horizon	<.5 <.5	<.5	< ، 5
	B horizon	<.5	<.5	<.6
Radium pc/gm	A horizon	.83 .61	.37	.48
	B horizon	.60 .34	.34	.36

points on the Faraday east line (see Figure 21). These points overlie gabbro; pegmatites and ore zones are further downhill to the south. The B horizon values of radium and uranium are in the range given above. Values for the A horizon are slightly higher than those given above (radium values are 1.3 to 1.5 pc/gm, and uranium up to 1.0 ppm).

Data cited by Vinogradov (1959, Tables 121, 122 and 123) give some support to the radium background shown in Table 28. The A horizons of ten soils from the U.S.S.R. have radium contents ranging from 0.5 to 1.1 with a mean of 0.9 pc/gm. Eighty-six soils from different parts of the world, undifferentiated as to horizon, have radium values ranging from 0.1 to 3.8, with a mean of 1.2 pc/gm. Six soils sampled at various depths below 11 cm have radium values ranging from 0.22 to 1.1 pc/gm. The background expressed in Table 28 is consistent with this data.

Uranium values cited in the same publication are substantially higher than those obtained at Monck Lake and at the north end of the Faraday east line. The A horizons of eight soils from the U.S.S.R. have uranium values with a range of 2.6 to 4.0 and a mean of 2.8 ppm. One soil sampled at 40 cm and lower has uranium ranging from 2.4 to 2.7 ppm. The lack of agreement between the Bancroft data and that cited by Vinogradov is surprising in view of the variety of soil types which he considered.

On the basis of the preceding four paragraphs, the radium background in the Bancroft region can be considered to be well established at 0.4 to 0.8 pc/gm for the A horizon and 0.3 to 0.6 pc/gm for the B. Uranium background is not well established, but can be assumed with reservations to be less than 1 ppm in both horizons.

Results of radium and uranium determinations in soils are shown

in Figure 5. Results for soils over mines are presented in the form of logarithmic histograms. Regional backgrounds are shown by arrows, double-ended for radium and single-ended for uranium.

Four different methods of prospecting are provided by the two elements in the two soil horizons. In all four cases, most of the values over mines are higher than background. Evidently all four methods may be useful in intermediate prospecting.

CHAPTER 10 SUMMARY

GENERAL CONCLUSIONS

- 1. A rapid analytical method has been developed for measuring radium-226 in sediment and soil. The sample is immersed in water in a sealed bottle and, after several days equilibrating time, nascent radon-222 in the water is measured. The amount of radium is calculated by dividing the amount of radon by $1-e^{-\lambda t}$ where λ is the decay constant for radon and t is the equilibrating time. Other sources of radioactivity such as the thorium decay series and potassium do not interfere.
- 2. Using a single radon measuring apparatus, one person can make 20 to 25 radium determinations daily; however, one person could operate two or more apparatus and achieve an output of greater than 50 per day.
- 3. Accuracy and precision of the method are adequate for geochemical prospecting.
- 4. The detection limit is 0.02 picocuries per gram, but no samples were below the limit.
- An alternate method, in which samples are stored dry and radon builds up in brass tubes gives superior precision. It appears to be simpler but, because only a few samples were treated, its productivity cannot be assesed.
- 6. Radon emanation is less efficient from dry sediment than when the sample is immersed in water, that is, a larger proportion of radon produced escapes from wet than from dry sediment.
- 7. Combined sampling and analytical errors for radon in water and for radium and uranium in sediment and in A and B horizons of soil are

low compared to the range found in nature. Uranium in water is a useful prospecting tool in spite of its high sampling error relative to natural range.

- 8. The advantage of low analytical error for radon in water is destroyed by the high sampling error time.
- 9. The sampling error for radium in the A horizon of soil is lower than for radium in the B and for uranium in the A hroizons.
- 10. Radium in surface water is generally below the practical lower detection limit of 1 pc/1.
- 11. Under conditions prevailing at Bancroft and under the assumption that contamination has been recognized, radon and uranium in lake and stream water and radium and uranium in organic and clastic sediments are all useful in reconnaissance geochemical prospecting for uranium. With the possible exception of radon and uranium in lake water, all of these are useful in detailed prospecting. Methods using sediment show a more accurate response to uranium deposits than do methods using water.
- 12. Radium and uranium show a more accurate response to uranium deposits in clastic than in organic sediments.
- 13. Data for clastic and organic sediments can be plotted on the same map by applying a correction factor.
- 14. In drainage sediment surveys, radium and uranium show equally accurate response to uranium deposits.
- 15. In detailed prospecting using soils, radium and uranium in the B horizon and radium in the A are equally useful, but uranium in the A horizon is of no use.
- 16. Radium and uranium in both horizons are useful in intermediate prospecting.
- 17. Radium-226 is enriched relative to uranium in clastic sediments.

has been precipitated.

- 18. Uranium is depleted relative to radium-226 in weathered rocks. This indicates that uranium is more thoroughly leached than is radium.
- 19. Radon-222 disappears exponentially with a half-life of 16 hours from one-foot deep, still water exposed to the atmosphere.
- 20. Radon in surface water and groundwater generally has a local source, within a few hundred feet. In other words, radon is immobile.
- 21. Radium-226 in surface water at Bancroft is not sufficient to account for the radon-222.
- 22. Radon below the water table does not, in general, move as a gas.
- 23. Radium-226 in sediments in the Bancroft area is high enough, on the average, to account for all the radon-222 in water.
- A model is proposed in which the level of radon-222 in surface water is controlled by addition of radon through decay of radium-226 in the sediments and influx of radon-charged groundwater, and by loss of radon due to aeration and radioactive decay. Results of stepwise multiple linear regression analysis of data collected for 182 stream sample points lend strong support to this model.
- 25. Simple correlation coefficients are less useful than stepwise multiple linear regression in evaluating this model.
- 26. In addition to supporting the above model, the results of stepwise multiple linear regression show that, other things being equal, the level of radon-222 in water is inversely related to the amount of organic material in the sediment. Evidently organic material reduces the radon emanation efficiency of the sediment.
- 27. Radium and uranium are concentrated in organic relative to clastic sediments. Sediment samples containing plant parts have radium

and uranium levels intermediate between those of organic and clastic sediments.

- 28. Radon is low in lake and pond water, intermediate in stream water and high in spring water.
- 29. Lognormal distributions are exhibited by radium and uranium in sediment and by radon in water.
- 30. Radium is higher in the A than the B horizon of soil. Uranium is about the same in both horizons or slightly higher in the B.
- 31. Radium and uranium are both precipitated from surface water by organic matter. Precipitation of uranium is much more thorough than radium.
- 32. None of the element distributions considered, radium and uranium in sediment and radon and uranium in water, are correlated with the season.
- 33. The level of radon in surface water is not correlated with daily rainfall.
- 34. Daily level of radon in a control lake is not correlated with that in a control stream.
- 35. Correlation between the level of radon and turbulence in surface water is weak or nonexistent.
- 36. Seasonal change in flow volume measured for a control stream is not, or very weakly, correlated with the level of radon in the water.
- 37. Radium and uranium are enriched in fine relative to coarse fractions of clastic sediments.

RECOMMENDATIONS FOR GEOCHEMICAL PROSPECTING

Recommendations for geochemical prospecting for uranium are summarized in Table 29. Material to collect and elements to determine are indicated for three sampling densities.

At the early reconnaissance stage (less than 1 sample per 2 square miles), drainage surveys should be used. Sediments give better results, but lake waters are often cheaper to collect. Clastic sediments should be collected wherever possible and organic sediments otherwise. A correction must be applied before showing results for organic and clastic sediments on the same map. Either radium or uranium can be used, with equal success. If waters are used, the choice between radon and uranium should be based on logistics. If a small lab can be set up in the field, then radon can be used; otherwise uranium must be used.

At the later intermediate stage (0.5 to 200 samples per square mile), sediments are preferred. If rapid analytical feedback is imperative, then stream or pond waters can be collected and analyzed for radon. If drainage is lacking, then the A or B horizon of soils can be used. In sediment and soil surveys at this scale, radium and uranium are both useful. Clastic sediments are more useful than are organic.

At the detailed prospecting stage (more than 20 samples per square mile), sampling is denser than the drainage system allows. The B horizon of soils may be analyzed for radium or uranium. If the A horizon is sampled, then only radium can be used. Soil gas can be analyzed for radon at the sample location (Dyck, 1968), but the results of this technique have not been compared with the methods considered here. Uranium in plant parts has been used (Hawkes and Webb, 1962, p. 376).

The use of radium in plant parts is suggested but has not been tried. If weathered rock is collected, and if the uranium mineral are older than about a million years, then radium analyses are highly preferable to uranium.

SUGGESTIONS FOR FURTHER RESEARCH

- (1) The mode of transport, chemical or physical, of radium in drainage can be determined by analyzing heavy mineral separates for radium, and then, if radium is not transported in heavy mineral grains, making an autoradiograph of a section of a grain mount. A concentration of radioactivity on grain surfaces would indicate that radium is transported chemically and then precipitated.
- (2) Gamma spectrometry might be more useful in analyzing for radium than is the method presented here. If radium in sediment can be detected by this method, then carrying a portable gamma spectrometer along streams might be an effective method of prospecting.
- (3) Results of geochemical surveys such as those presented here should be compared with results of geophysical surveys such as ground and airborne gamma—spectrometry, to determine which method is more effective and efficient.
- (4) The biogeochemistry of radium applied to prospecting for uranium should be investigated.
- (5) Application of the other members of the two uranium decay series, as well as the fission products of uranium, to geochemical prospecting for uranium is worth considering.

TABLE 29
Recommendations for Geochemical Prospecting

Scale of Survey and Sampling Density	Mater	tal Sampled	Element Determined	
Reconnaissance <1 sample/2 sq. mi.	Sediments preferably if they can be collect— ed cheaply	Clastic preferably Organic if no clastic	Radium or uranium Radium or uranium	
		I sampling by	Radon if a small field lab possible Uranium if not	
		Clastic preferably	Radium or uranium	
Intermediate	Sediments	Organic if no clastic	Radium or uranium	
0.5-200samples per sq. ml.	0 - 21 - 2 - 3	id analytical imperative	Radon	
		on of soil if no nsity greater than samples	Radium or uranium	
	B hor:	0.2 (6.2 %) Sec. 2 %	Radium or uranium	
Detailed >200samples/sq. mi.	Soil A hor:	izon gas	Radium Radon	
	Plants		Uranium has been used Suggest try radium	
	Weathered roo	ok.	Radium if older than 1 million years; uranium if younger	

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APPENDIX I

NATURAL RADIOACTIVITY

The material of this section is found in many textbooks on nuclear physics and nuclear chemistry, for example, Kaplan (1962).

The rate of transmutation of a radioactive nuclide is proportional to the amount of the nuclide present, thus:

$$\frac{dN_1}{dt} = -\lambda N_1$$

where N_1 is the number of atoms of the parent nuclide, t is the time and λ is a decay constant in units of $\frac{1}{t}$ unique for each radionuclide. The solution of this differential equation is

$$N_1 = N_0 e^{-\lambda t}$$

or

$$N_0 = \frac{N_1}{e^{-\lambda t}} \qquad \dots (1)$$

where N_0 is the number of atoms of parent present at t=0. The halflife or amount of time for half of the parent to decay is $\frac{\ln 2}{\lambda}$.

The daughter species forms at the rate at which the parent decays, $\lambda_1 N_1$, where λ_1 is the decay constant of the parent. If the daughter is radioactive it decays at the rate $\lambda_2 N_2$, where λ_2 is the decay constant of the daughter and N_2 is the number of atoms of the daughter. Thus:

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 .$$

The solution of this differential equation is

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_0 (e^{-\lambda_1 t} - e^{-\lambda_2 t})$$
 ...(2)

assuming no daughter was present initially.

If the parent is so long lived that it doesn't change appreciably during the experiment, $e^{-\lambda_1 t}$ is close to 1 and, furthermore, if the daughter is very short lived relative to the parent, $\lambda_2 >> \lambda_1$, and (2) reduces to

$$N_2 = \frac{\lambda_1}{\lambda_2} N_0 (1 - e^{-\lambda_2 t})$$

or

$$N_0 \lambda_1 = \frac{N_2 \lambda_2}{1 - e^{-\lambda_2 t}} \cdot \dots (3)$$

After a certain time $e^{-\lambda_2 t}$ becomes close to zero so that

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$$N_2\lambda_2 = N_0\lambda_1$$
.

In other words, the rate of decay of the parent is equal to the rate of decay of the daughter. This situation is known as radioactive equilibrium. Radioactive equilibrium is approached with the half-life of the daughter. Starting with pure parent, daughter activity is equal to one-half parent activity after one daughter half-life, three quarters after two half-lives, etc. Radon-222 approaches equilibrium with radium-226 asymptotically in a few weeks.

APPENDIX II

ERROR THEORY

Analytical error or precision is determined by running a set of samples more than once each and is expressed in terms of the standard deviation s. The formula is given by Wilson (1952, p. 245):

$$\varepsilon = \sqrt{\frac{\sum_{j=1}^{\Sigma} \sum_{i=1}^{(X_{i,j} - \overline{X_{j}})^{2}}{\sum_{N-k}}}$$

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where k samples were determined n_j times each, X_{ij} is the ith determination of the jth sample, $\overline{X_j}$ is the mean of the jth sample and

$$N = \sum_{j=1}^{k} n_{j}.$$

For trace elements, which have an approximately lognormal distribution, absolute errors associated with high samples are much higher than those associated with low samples: relative errors should therefore be used. This is done by dividing each deviation from the mean by the mean for that sample. Thus:

$$s = \sqrt{\frac{k}{\sum_{j=1}^{\Sigma} \frac{\Sigma_{ij} - \overline{X_{j}}}{\overline{X_{j}}}^{2}}}$$

If each sample is analyzed only twice, this reduces to

$$s = \sqrt{\frac{2 \sum_{j=1}^{k} \frac{X_{1j} - X_{2j}}{X_{1j} + X_{2j}}^2}{\frac{1}{k}}}$$

where k is the number of pairs of analyses.

Relative errors also may vary over the range of concentrations, for example, higher relative analytical errors are commonly associated with lower concentrations. However, if the replicates are selected at random over the range of concentrations considered, the above formula will give an unbiased estimate of the standard deviation.

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APPENDIX III

PROCEDURE FOR MEASURING RADIUM-226 IN SEDIMENT AND SOIL

Weigh 40 ml of sample (-10 mesh) into a numbered bottle. Add about 50 ml water which is low in radium and radon. Replace lid and shake vigorously to wet the sample. Fill with water. Record time and date. Let sit several days.

The next three steps are identical to those described by Dyck (1969a, p. 29):

Switch counter on and let it warm up for about 2 minutes before making measurements.

Evacuate cell and admit air to cell through drierite trap. (Make sure that drierite is not used up.)

Place cell on photomultiplier tube after checking if pm tube and cell window are clean. Put cap on pm tube and engage high voltage switch. Wait one minute for the deexcitation of light in the cell with counting switch on. Zero scaler and timer and count "background" for 4 to 5 minutes.

Open bottle and pour out 50 ml of water to make room for bubbling. Install stopper with gas dispersion tube. Close both stopcocks, shake vigorously and connect to vacuum line.

Evacuate cell and close valve to pump. Slowly open outlet and then inlet valves to achieve a slow bubbling rate so that pressure in the vacuum line reaches atmospheric in eight minutes. Detach cell, place it on pm tube, and replace light shield. Start counter and observe the count rate level off as the light de-excites. This may take a minute or two. Again Dyck's (1969a) procedure is appropriate:

Exactly ten minutes after the start of the filling procedure zero the scaler and timer and take two five-minute counts.

Record date, sample number, time of day, cell number, background counts/minutes, sample counts/minutes.

Immediately after completion of counting of the sample, evacuate the cell and flush three times with air by filling and evacuating the cell.

Sample calculation

Weight = 25.0 gm

Water added: 10:00 AM, August 10

Count started: 12:00 Noon, August 20

Equilibrating time = 242 hours

 $1 - e^{-\lambda t} = .839 \quad \text{(Appendix IV)}$

Background: 3 counts/5 minutes = 0.6 cpm

Sample count: 15 counts/5 minutes

31 counts/10 minutes = 3.1 cpm

Net count rate: 3.1 - 0.6 = 2.5 cpm

Radium = $\frac{1.16 \times 2.5}{25.0 \times .839}$ = 0.14 pc/gm

APPENDIX IV TABLE OF $1 - e^{-\lambda t}$ FOR RADON-222

Time in hours followed by $1 - e^{-\lambda t}$.

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€ .	G.C443	7 0	.0515	- 3	C.C586		0.0657	1 C	0.0727
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21	C.1466	22 C	1.1530	23	C.1594	24	0,1657	25	0.1720
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36	0,2380	37 0	2437	38	C.2494	39	C.2951	4 C	0.2607
41.	0.2662	42 0	.2717	43	C.2772	44	0.2827	45	0.2881
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APPENDIX V

LIST OF DRAINAGE DATA COLLECTED

This appendix lists data collected for each of the drainage sample points shown on the sample location map (Appendix VI). Sample numbers are abbreviated by dropping the number 31. Data for each sample location occupies three lines: the first line for the water sample (W), the second and third for the sediment samples (S) at the same location. Where only one sediment sample was collected, the third line is blank.

Several of the sample points shown on the map represent more than one sample set. Location of these duplicate samples can be found by reference to the following table.

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Location on map	Sample numbers	Location on map	Sample numbers
31D16 8040	3004*, 4040	31F4 8141	4141, 4142
31D16 8155	3186 , 4155	31F4 8135	4135, 4136, 3685
31D16 8058	4058, 4059	31F4 11789	3789, 3595*
31D16 8165	3183, 4165	31F4 8067	4067, 4068
31E1 11163	3163 , 3025*	31F4 8069	4069, 4070
31E1 8100	4100, 3174, 3026*	31F4 8169	4169, 3408
31E1 11159	3159 , 3027	31F4 8145	4145, 4146
31E1 8055	4055 , 4056	31174 11826	3826, 3545*
31E1 8071	4071, 4072	31F4 8120	4120, 4122
31F4 8073	4073, 4074	31F4 11667	3867, 3530*
31F4 11714	3714 , 3534*	31F4 8030	4030, 4031
31F4 11774	3774, 3548*	31F4 8032	4032, 3533*
31F4 8180	4180 , 3677	31F4 8021	4021, 4022, 3800
31F4 8181	4181 , 3680	31F4 8027	4027, 3532
31F4 11766	3766, 3546*	31F4 8082	4082, 3531
31F4 11769	3769, 3547*	31013 11157	3157, 3001*
31F4 11792	3792 , 3591*	31013 11162	3162, 3002*
31F4 8137	4137, 4138	31013 8037	4037, 4038
31F4 8139	4139, 4140	31013 8105	4105, 3300
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*Water samples only. Not included in list of data.

Location of sediment samples are not shown on the map (Appendix V), but can be found by reference to water sample numbers. Samples collected in 1969 (numbers above 4000) are listed in the first part of the table in numerical order. Samples collected in 1968 (numbers from 3000 to 4000) are listed later in alphabetical order. Sample points 4177 to 4179 are off the map on the Crowe River.

An explanation of each column follows:

Sample number: followed by (W) for water samples and (S) for sediment samples.

Element concentrations: radon-222 content of water (W) pc/l, and radium -226 content of sediment (S) pc/gm followed by uranium content of water (W) ppb and sediment (S) ppm (-l indicates no analysis).

Type of drainage sampled: 1 = stream, 2 = pond, 3 = swamp, 4 = 1 ake, 5 = spring and 6 = drill hole.

Width: of water body in feet for streams, ponds, swamps and springs.

Area in square kilometers for lakes.

Depth: in feet at sample point.

Vegetation (3 columns): First two columns are in Geological Survey of Canada code. Third indicates intensity of tree cover: 0 = open, 1 = sparse, 2 = moderate, 3 = well wooded.

A: water level: 1 = dry, 2 = low, 3 = normal, 4 = high, 5 = flood.

B: rate: see page

C: turbulence: see page

D: precipitate: 0 = none, 1 = Fe, 2 = Mn.

E: sample position: 0 = right bank, 1 = center of stream, 2 = left bank, 3 = shore of lake, 4 = center of lake.

- F: water color: 0 = clear, 1 = white, 2 = yellow, 3 = orange, 4 = red, 5 = brown, 6 = black, 7 = green.
- G: sediment color: 0 = white, 1 = buff, 2 = yellow, 3 = orange, 4 = pink, 5 = red, 6 = brown, 7 = dark brown, 8 = black, 9 = grey.
- Composition of sediment (6 columns): First column is gravel, second is coarse sand, third is fine sand, fourth is silt, fifth is clay, sixth is organic material. Entry indicates proportion of that material in tenths.
- Type of gravel fragments (GRAV): Geological Survey of Canada mnemonic code.
- Contamination: refers to contamination evident at sample location: 1 = none, 2 = possible, 3 = probable, 4 = definite.

Temperature of water.

pH of water.

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                                                 N M
          (S)RADIUM URAN. PT T E
                                        MENTS V T P
           . PC/GM PPM E H H T ABCDEFG
 D 16-4165(W) 67 0.9 1 18 122A23100106125 2GRNT12468
 D 16-5) 65(S) 0.423 7.80 1.0 1 6127
 E01-6165(S) 2.262 10.20 .3
                                      2 7 1.9
 F04-4166(8) 36 0.5 1 10 032P33122106244 GRNT11871
 F04-5166(S) 0.466 3.00
                                     1 6244
 F04-4167(N) 25 1.7 1 2 012E23220106424 GRNT12074
 F04-5167(3) 0.386 2.60 2 6 28
 F04-4168(%) 5 0.5 1.70 2P33310006 91 2 69 F04-5168($) 0.186 1.10 03 2 6 91
 F04-4169(8) 88 1.2 1 5 032E23100126 44 2
 F04-5169(S) 2.410 8.20
                                 1 6 44
 F04-6169(S) 42.968 55.00
                                     0 7
 E94-4170(X) 170 2.2 1 2 033 23110126 8 2 1 64
                                     1691
 F04-5176(S) 6.177 16.60
 F04-6170(S) 8.177 15.60
F04-6170(S) 39.352 50.00
                                     ö_7____
 F04-4171(3) 146 2.4 1 3 083 23110129
 F04-5171(8) 5.873 10.60
E04-4172(V) 496 0.3 1 1 012P22111106334 GRNT11654
E04-5172(S) 0.240 3.60 1 6334
 E04-4173(4) 118 2.2 1 1 0,62P23100128 1 9GRNT1
 F04-5173(S) 2.108 11.00
                                    1 6136
 #04-6173(S) L8.508122.00
                                     1 8 1
 F04-4174(%) 116 0.6 1 3 42P2310 127
F04-5174(%) 2.582 9.00 1 7
                                           19
 F04-5374(S)
             2.582
 E04-4175(3) 45 0.6 3120 2P2300 326
E04-5175(8) 4.735 7.60 1.0 3 2
                                           19
 F04-4176(图)
                   2.0 1 10 102P23100129
               285
 E04-5176(5) 20.589 52.00 20 1 8

E04-6176(5) 10.552 35.00 20 1 9 9
                                     199
              14 0.5 1 45 303 332120062 7
 C13-4177(R)
 C13-5177(S) 1.342 0.76 .5 0 62 7
 C12-4178(N) 1 0.5 1110 2E13200207 8 2 C12-5178(S) 0.556 0.48 1.0 2 7 8 2
             0 0.5 1.75 2E23222106343 GRNT12174
 C12-4179(W)
             0.552
                   0.64
                          1 6343
 C12-5179(S)
              9 0.5 1 3 023 23110106127
                                            GRATLI877
- FO4~63.80(反)。
             0.281
-- E04~5180(SI
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afilipera ang menjerang termini perang ang ang anaman ang manera na pelikang termini M	NUMBES SAMPLE	regional and a second	เกลน	10-8311-148	e na para di di	e grani. IJ	en merketa salam. V	agentina was say of the second	ſ	O MO I	N. G	С 3
	MUMBER	9(: /1	D D D	T	I	E E		****	OF	Ř	n E
And the second s	, a. 5 (a. 5 a. 5a. 72 5 726	entre de Sastanda en 1919. L	Silvino III Portures my a	on kana 1969 balan san mengan	Υ	Ū	P G		S	EDI.	- Д	
		(5)0/	MUTCH	URAN.	P	Ţ	T E		! -	ENT	S V	Ϋ́P
		þ(:/GM	ррм	E		H T					A PH
aligada da king karang manakan dan dan dan persamanan manakan penjangan menjangan menjangan menjangan menjang	مندون ويعاورون والمشارة	ente je oje od aj romor	e Se posta e de la Seconda de	ر ماندي موار از د د دارا يون پيدمندي دي هاد	ogy navertner	re, consider provide	Line - I may come and a	. (4	reconstruction engines		may decide the	M
	4-4181(12169
Fo:	(-518) (<u> </u>),385	3.00	ti ili sanda		25		0 65	34	<u>, , , , , , , , , , , , , , , , , , , </u>	w
			2040	4 6	,	•	AT-AU	3.5 1 1	100		1.0	21744
and and a series of the control of t	4-41821	:智). - なくえる i	3240 330	1.0	1		OTCH	5517	1 8 1 8		1.9 1.4	3176.6
	1-51821	54 4 J	しゃりかけま	(46.40E			. ;		r o		1. 7	
etalen system in die materialiste in der en en de de en	1-4193(rentenskaper (d.)		0.5	4	re refetatatel	211	33000	306	46	pagina telakoronia en	1246.1
	l-5183(2,
		ade a antique est de la company de la co	raisteile.	and Technique	er erber	- v. 111 - 21			_ ` `			
C1	3-31471	A.).	O	0.0	4	2	28	33000	3061	27	GRNT	1238.7
G 1:	3-3149(S) ().067	0.54	A-5		0.3		3 61	27		
رای در این از در این از در این از در این	Na kasantahan basar atau sama	ومادر شواهیمان و در درستان	e dan katalan katalan sa	enter of distributions and the control of the contr	· • · · · / • · · · · · · · ·	مني بدوجون المساور المساور	and the same of the same of the same of	والمراجع والمراجع والمراجع المراجع	erie, generalis et e e		and the second second second second second second	فالمان والمراجع معالم
C 1	3-3149(W.)	31	O * O	1	1.1	0429	33100			19	1.167.7
<u>C1</u>	3-3150(-S.)	660	0.48			0.6.		0 7) 9	
			<i>r</i> 5	Λ 1	<u>, , , , , , , , , , , , , , , , , , , </u>	1 /.	3	22010	14.07	7	3	12480
aj mana se potentiales, esperante trenen a montre de despres	3-3151(3-3152(. W.J.)	7. 20		7 E	270 270	22010	4 7		3	1 Z TOO
) 27 34 344	34 J. 18 S.).*⊙O.a.	ft #.Z. W		-	3.1423		<u>-</u> ₹ 1	•	٠,	
	3-31531	94	22	(), 3	4	14	· · · · · · · · · · · · · · · · · · ·	33010	406	er og er e veze veze e	19	12580
	3-3154(
	y Nguy Tin Pilatina a di Ta	Miringer Welf	kati yi ta ad Qiya	a, formalismos per en e			we Proceeds					
c 1	3-31571	14)	1	0.4	1	20	1.32E	23100	0006	1	9	.11879
C1	3-31581	5) 4	3.322	0.60			1.0		0 6	}.	9	
	Geografia de la completa de la comp		ر د در ده و خو سرمانه کرد. و در د	fastaniania yazori ng wo			and the second	gerger gebieden.	77		, <u>,</u> ,,,	71.500.3
CI	3-31.59(分)	233	35.0	3	30	253	23100				11881
C1	3-3160(3-3161(/ 19Ω - 728 -	3.00			. <u>6</u> 9		$\frac{1}{1} \frac{6}{8}$		19	
C.A.	5-3301 (3-3162 (- 511.11 - 541	1000 K		1	26	093	23102				11678
	377 D J G C J 377 D J G C J	C1120	1 37 7	78.00		2	0.9		1 6		19	
i i	3-3165(3-3164(\$} 23	3.373	8.20			ĊЭ		2 84	33		
ČÎ	3-3 <u>1</u> .65 (W}	156	25.5	1	10	052E	32211	1063	222	LGRN	11885
Ć).	3-31660	\$1 3.4	4.970	3.30			0.5		2 63	232	A	
											0.54.5	r
c_1	3-3167(W)	238	0.8	1	40	1.025	32002	22063	331	GRIV	11.686
C1	3-3168(\$} '	1.797	11.40			1.0		2 63	331		
and the state of the	and the second of the	and a management of the second	paga ang kang kang kang kananang kananan Langga kang kang kang kang kang kang kang	÷1:.0	1.4	. 3		22000	12071	35	10000	112188
C.	3-3170(₩] -61 /	W. 654 (= 3	— (0 a (0 − - (0 a (1 c) − −	体以	. 60	0.4.	73 72 63 C	3 71	35	1	3.22.00
	3-3171 (3-3172 (000	0.34	es alongia		06		3 6		19	are constant
	5 5 1 7 2 1	1.5 %	ì	0.2	1	12	183	33100)	26	1.678191	13,207,7
	2-21741	ं	.03Î	0.22		. M. 67 . 4	18	". i	1 71	26	į	
ř.	3-3174(3-3174(S)	1.152	0.62			1.0	na artist	1 7	na wha she who	19	e e a grande de la compansión de la compan
Ci:	3-31791	W.)	19	0.1]	3	0.33	33000	,,			11463
ca.	3-3180(8): (8	239	2.40			0.3		.1. 6		19	
又 真其的。		· + ,		, .				2200	0.07		; ; ; ; ; ; ; ; ; ; ; ; ; ; ; ; ; ; ;	12272
y (C1)	3-3191(W)	1	0.1	4()	• 2	40	5500	3 6		19 19	183.43
C1.	3-3182(5) (3.4号等。	30.00			4.0		. O		4 4	
						1						

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SAMPLE LAIRADON URANIUM W D V
                                      COMPN. G C T
                      Y D P G
                                      OF R
SEDI- A
  NUMBER PC/L
                                               0, 6
                                               N M
        (S)RADIUM URAN, PT T E
                                      MENTS V T P
                 PPM E H H T ABCDEFG
           PC/GM
C13-3183(W) 18 0.2 1 25 0.73 23211106432 1GRNT11776
                  0.92 07 6 64
C13-3184(S)
            9.445
                                   2 8 19
C13-3285(S)
           0.634
                 3.20
                  -1.0 30.1 022833000126 1 9
C13-3191(W)
G13-3192(S) 0.297 0.28
                                        19
                           02
                                   1 6
0.13-31.93(5) 0.103 0.06 02 1.6.11 8 0.13-31.94(4) 54 \pm 1.0 1 2 0.33 33112107222 4GRNT11780
C13-3195(S) 0.136 0.36 0.3 1 6333 1
           0.282
                 0.68
Cl3-3136(S)
                        - 0.3
                                   1 7
                                        19
                  0.2 4 1.4 2E2300
C13-3300(W)
            . 5
                                               2237.9
C13-3301(S) 1.931 7.00
C13-3302(S) 0.206 2.60
                                   3 6
                                   3 6127
0.16 + 31.55(\%) 22 0.3 4 14 503 33000407 19
                                               12280
D16-3156(S) 33.300 42.00
                                         19
016-3157(W) 33 0.1 1 30 303 33000107
                                        19 1217.1
·016-3158(S) 1.338 3.00 30
                                   1 7
                                         19
D16-3159(8) 757 3.0 1250 352P33100108 2 8
016-3160(S) 62,700 50,00 35 1 8 2 8
             364 0.7 4 14 603 33010407
り136-3162(3)
                                   4 7
Di6-3163($)264.000 32.00
                       60
D)6-3154(W) 28 0.0 4 1410.03 33010407
                                         19
                                               12380
016-3165($)205.171 22.00 100 4 7
                                         19
018-3147(1) 10 0.0 4 14 603 330:0407
                                         19
                                               12276
D16-3158(S) 16.570 18.20
D16-3159(W) 17 0.1 4 14 3 33010407
                                         1.9
                                               1227,8
D16-3170(S) 17.346 5.00 9.0
016-3171(%) 6 0.3 4 14 3 3301 407
016-3172(S) 57.670 16.70 17.0 4 7
                                         19
                                               1227.8
D16-3173(W) 27 0.3 4 14 3 33000407
                                         19
016-3175151104.653 38.00 70
                                         19
                                   4 7
                                         19
016-3174151 78.274 16.60
19
                                              1,237,8
016-3173($) 13.308 6.70
            4 0.2 4 14 3 330103061 36 GRNT12480
D16-3179(W)
                          20 3 61 36
D16-3190($) 10.035 1.66
                 0.2 4 14 3 330003061 9 GRNT12480
D16=3181(W): 17
                          1.0
Dia-3192(5) 4.020
                  0.74
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			and the second seco				G		SEDI-		NM
Land Color, Life the Color Super Color Color Color	sar ou mountaine.	(\$) PAD1U								S V	T P A PH
		PC/GM	Яgq	C	11	in .	I ABC	# C	•		M PH
- 4 3 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4	016-3183(3 7	0.9		18		3 3310	0107	5	5	1237.4
1. 	016-31841	5) 4.26	2 14.60		2. ()	1.3		1 8		L 9	454
a tanan kana sara nasa tahun kana menggalan berangan di mengan mengan mengan mengan mengan mengan mengan menga Tanah sarah sa	D)6-3185(\$1 0.44	3 4,40	V 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2		0.7	en e	2 6	9	1	
	016-31866	W) 2	8 1.1],), 5	2,53	3 3310	0006	9	1	11977
N	016-3187(!		1.5		0 6	9	1	
tarili kan	en de la companya de La companya de la co	e rengiographical construction in renormality		i reserve	de de la companya de						
	D16-3215(11,57.2
a and a state of the same of t	D16-3216(21 0+41	3 - 4.23			, Çatı	one of the second	. <u></u>			
	661-31111	W)	0 1 3	4	2		3 33000	0406	.]	19	1215,8
y <u>Mangapagana an</u> arawa a kata arawa a	E01-3111(E01-3112 (5) 0.374	2,20			4.0		4 6		9	
nde Blackers of and est											
a. Mangalaga kan mengabbangkan galaga kan kembanah ang menang ang mengan mengan pang memba	E01-3113(12160
	E01-3114(\$) 0.73	B 3.00	i ,,		5.00		4 7		l 9	
	. † 03 - 019 til	2.9 6	o /\ \	1.	2	•	2 22017	2206	261	1 00 80	(222KB
alahahan ang an sing sagir bata.	E01-3115(*) **	5 0.0 1 7.80	/ / } !	6.	ΛT.	5K 2 \r i.	7500	1321 1351	Tistana.	Z 2.3 3.00
	TEMPART GALANT	.a.fa.f ⊆ 2	£ 1000			17.0			, , , , ,,	,A.	
eri dala galamika da kanasarah ke da kanasarah ke da kanasarah salah da kanasarah salah da kanasarah salah da k	F01-3151(W	1 0.0	1 1	3	0.5	3 2321	0126	261	1	1237.4
	E01-3152(\$1 0.16	0 2.20	}		9.5		1 6	361		
A STATE OF THE STA	F01-3153(5) 0.93	8 -1.00			0.2		2. 1		9	
والمراجع والمراجع والمراجع والمستران والمستران والمستران والمستران	E01-3154(4 0.0		1	()4;	3 2421	1.1.27	1		1227.3
	E01-3165(5) 1.41	4 -1.00			()4		1. 7]	[9	
ega maga maga ka samatan saja peni da ka sa sa kalan da ka da da da ka sa sa ka sa sa ka sa sa ka sa sa ka sa		111	0.5	engara serengan		01	3 2322	2126	37	2C 0 N1	11570
	E01-3156() <u>6</u> 366			ZONT	1. 1. 2 1. 2
	E01-3152(s) 3.52	2 2.40 4 2.60	., : , l		0.1		\tilde{z} 7		9	
	E01-3159(9 0.0			0.10	3 3210	0128	1		12475
alitation of the sold of the state of the state of the sold of the	F01-3150(2 3.20			OI	to a second	1 8	,	[9	
er e	E01-3151(7 -1.00	جمعر پرمواد سا	ا معروضا مساوية	O(1)		1 8		1.9	name and an
The state of the s	E01-3163(왕) 2			5		3 3310				11874
<u> </u>	E01-31661	5) 3.3/	3 0.74			0.4		$\frac{1}{1}$ 8		ĺὸ r	
•	E01-3165(7 3.00 4 6 0	1	10.	LOS	2310	1227	, <u>, , , , , , , , , , , , , , , , , , </u>	ur 72GRNI	12064
er gegyn ei general er general fan stêre fan ei en en en en en. Op stêre fan en	E01-3167(The first control of the second	3 1.78	* .	2.5	03		2 6	î (}	A. C. C.
	E01-3158(:			2 7		9	
and the second of the second s	E01-31691	W) 125	9 34.0	1	1	1.00	2M3310	01.07	24	4	41366
and the second s	E01-31701 E01-31711	51 27.72	9480.00			0.0		.2.6	37	ere e	
			7174.00			1.0) 7	2	8	357/0
e de la companya de	F01-31721		5 0.7			0.31	3_3210	0127	7 25 7 25	3	11768
	F01-31731	5) 6.45	a 14.00			0.3		j. /	63)	
dialogo de Loberto, respekçita (s. 1811).		generally seem of the entire terms of the enti	3 0.3	1		773	3 2310	กับกั	,	9GRN1	1207.2
	E01-31741 101-31751							1.7		9	are see to 17.55
) *	A 44 4 7 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1										
	E01-31781	(2) 5	3 0.4	j.	4	0.52	2F3310	0177	7	3	12071
	£01=31771	5) 0.63	2 2.40			0.5	:	1 7	8		
hilingi (dhambara e amendada)	F01-3178(5) 1,12	7.3.60	La esta esta esta esta esta esta esta est	and the state of t	<u>. 45</u>	para di kacamatan	1 2	A CONTRACTOR	(9	er to security of the control

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SAMPLE IMIRAGON URANIUM W D V
                                       COMPN. G C T
  NUMBER PC/L PPB TIEE
                                       OF R OE
                      Y D P G
                                       SEDI- A N M
                                      MENTS V
        (S) RADIUM URAN. P. T. T. E.
                                                TP
           PC/GM PPM E H H T ABCOREC
E01-3179(W) 51 1.0 1 15 152A23100127 19
E01-3180(S) 0.762 7.46 08 1 7 19
E01-3181(K) 38 0.1 1 1 012P33222127332 2GRNY11675
E01-3182(S) 0.516 3.00 01 1 6 64
E01-3183(S) 2.034 3.00 00 1 7
                                   1 7 19
                  0.0 1 2 0.22P33210106261 1GNSS11876
EO1-3184(8)
            105
                  4.60 02 1 6 91
EO1-3135(S) 1.289
                                    2 7 19
                 8,20
                            00
E01-3186(S)
            1.318
                  40.5 1 1 042M33111107222 4SCSY21678
            12120
EQ1-3192(4)
            7.537 92.00
6.124126.00
                                   1 7333 1
                            0.4
F01-3193(S)
                                   1 7111 7
                           0.4
E01-3194(S)
            17 -1.0 1 25 043 33102106541 MRBL11972
E01~3204(8)
ED1-3205(S) 0.256 4.60 04 1 6541
            37 -1.0 1 10 063 33100127222 45RNT12267
<u> 房价1~3208(以)</u>
E01-3209(S) 0.097 4.40 06
                                0 6343
£01-3210(S) 0.393 2.60
                           0,4
                                    1. 7.
                                           19
                                           10
            22 -1.0 1 50 052P33100107
E(6)1-3211(日)
E01-3212(5) 1,179 20.00 06 1 7 19
F04-3408(N) 119 1.2 1 4 062E23100126 8 2 12076
F04-3400(S) 5.340 26.00 06 1 6 91
F04-3410(S) 18.200 -1.00
F04-3419(W) 115 -1.0 4
                                F04-3420(S) 0.458 5.80
F04-3662(%) 3 0.4 40.1 2P33010306143 2GRNT2238.5

F04-3661(S) 0.219 1.80 3 61.54 2

F04-3662(S) 0.264 17.40 3 6 19

F04-3663(V) 93 1.4 1 5 0.13 2211210755 GRNT1197.5
E04-3654(S) 0.349 7.80 01 1 0755
                  0.6 1 3 0.52E32100106 45 1
F0%-3465(V)
F04-3566(S) 6.310
                            0.5
                                         45 1
                   0.84
                   0.0 1 1 052M33100107113 58CST11581
           43
E04-3567(9)
                   0.90 05 1 6226
            0.136
P04~3668(S)
F04-3869(S) 0.493 -1.00
E04-3676(N) 1 0.0 40.2 2033010306334
                                           MR8L12384
                                    3 6334
                  0.38
E04-3573($)
            0.171
                  0.4 1 1 022633110107 11 8
                                                11181
F04-3572(N) 120
                   1.40 1 02 1 1 1 1 8
            0.963
F94~3673(5)
             0 0.1 40.5 2433010306125 2MRBL12385
正の4~3674(夏)
                                    3 6136
                  1.34 3 6136
0.84 3 6 19
F04-3575(8) 0,068
£04~3676(S)
            0.095
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		PC/L		i i			OF R O E
			•	Y D	Р		SEDI- A N M
	talik partali Agging paratik aras di mas manan s	(2)89010W	the first the first of the property of the second states of the second s	A ** - * * * * * * * * * * * * * *	Ţ	e manera a ser escolar accesa y a sign	MENTS V T P
		BCACM	рьм	EH	Н	T ABCDEFG	H9 A
		يباريه بهاكروا والأوابية والمتحددة		فيح سنخوس		بأراب والوابية وأوالأ ويوا	
	04-3677(04-3678(243 IGRNT1169.1
	04-36791				0.2		
	04-36801					E331000071	19 21 6GRNT11775
the street of the second contract of the second	04-3631(the continue to an admitted the decision of the continue to the continue of th				1 63	
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F04-3850(S)	0.196	1.38			06		1. 7	19	·	

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Ontario Graduate Fellowships, 1967-68,

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"Radium geochemistry applied to prospecting for uranium", Can. Mining J., May, 1969

"Geochemical exploration for uranium: radium joins radon, uranium as indicators", Northern

Miner, March 5, 1970, p. 21

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using radium with those using radon and uranium", Third Int. Geoch. Expl. Sympos.,

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