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URANIUM PROSPECTING WITH ^{222}Rn IN FROZEN TERRAIN

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ABSTRACT

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Published reports show that ^{222}Rn contents of soil gas can increase under snow-covered or frozen soils. By utilizing these observations and results of field tests described here, it can be stated that U prospecting with ^{222}Rn in frozen terrain is practicable.

Rn profiles in frozen and snow-covered soils over U mineralization in the Bancroft area of Ontario outlined known radioactive zones more clearly than did scintillometer profiles.

Tests in Gatineau Park, Quebec, in the proximity of a radioactive pegmatite dike showed that lake ice acts as a restrictive barrier to Rn movement from lake waters beneath. Water samples, collected and allowed to freeze in plastic bottles, retained dissolved Rn quite effectively thus permitting sample collection and storage under the most severe winter conditions. Rn sampling of through-ice lake waters is therefore a feasible prospecting tool.

Samples of spring run-off (snow-melt) waters and slushy snow collected from within a known weakly radioactive zone near South March, Ontario, were shown to contain less Rn than found in the same stream waters in the summer. No pronounced Rn degassing event of frozen soils was apparent early in the spring thaw in percolating run-off waters draining from the zone. The usefulness of spring thaw hydrogeochemistry using Rn is discussed.

INTRODUCTION

The question of what is the fate of Rn in soils and waters in frozen terrain, especially in the Canadian Arctic, is important to U prospecting methodology. The mechanisms for Rn migration and escape to the atmosphere in the Arctic are probably the same as those operative in more temperate southern regions of Canada under severe winter conditions. The field tests described herein were designed to investigate such possible mechanisms and to measure the degree of accumulation of ^{222}Rn under both frozen ground and beneath thick lake ice cover in Ontario and Quebec under winter conditions. Earlier works on these problems and on the general consideration of Rn migration and dispersion in soils and other ground cover have been well reviewed by Tanner (1964).

Frozen ground, like wet ground, acts as a sealant tc Rn escape, resulting in an accumulation of Rn and its decay products at the frost line and in a decrease above the frost line relative to later frost-free conditions (Novikov and Kapkov, 1965; Khaykovich and Khalfin, 1961; Wennervirta and Kanranen, 1960; Alekseyev, 1957; Norinder et al., 1952; Kovach, 1945; Bender, 1934; Garrigue, 1935).

Garrigue (1934) also observed a 10,000-fold increase in Rn in the snow-covered dormitory of the Pic du Midi observatory compared with that found in summer air (Garrigue, 1951).

It can be seen that the increase in Rn activity which might be measured under winter field conditions would have to be weighed, in terms of value to U prospecting methodology, against the greater difficulties encountered in the field as a result of heavy snow cover, degree of ground frost or ice cover and generally uncomfortable working conditions.

Some further facts relevant to these considerations relate to the emanation characteristics of Rn into and from various geological materials.

The emanation efficiency of Rn from rocks and minerals into water was found to be about twice the emanation efficiency into air (Prutkina and Shashkin, 1967). Furthermore, the overall emanation from wet samples into air was observed to decrease relative to dry samples (Novikov and Kapkov, 1965); and wet, frozen samples held at −5°C emanated one-half to one-quarter of the Rn lost at 20°C. Thus Rn solubility and diffusion through ice and water are clearly important parameters in controlling the dispersion of Rn in surficial materials.

Such are the degrees of restriction on Rn mobility in frozen terrain, that in order to make optimum use of accumulations of trapped Rn at the frost line, it is necessary to sample quite close to this critical level.

Such sampling can obviously become quite difficult in situations where frost penetration is more than a half metre. Similar technical problems would likely be encountered in sampling lake waters beneath thick lake ice cover.

Nonetheless, under certain favourable conditions, winter sampling for ^{222}Rn can be regarded as a useful prospecting aid. Some such situations are described here.

RADON PROFILES IN FROZEN, SNOW-COVERED SOIL

In February and March, 1970, soil gases were sampled and tested for their ^{222}Rn content on U prospect located near the northwest corner of Faraday Township, Ontario. All of the work was carried out when there was a minimum of 60 cm of snow on the ground. The first tests were conducted over diamond drill holes that were known to contain U mineralization. Subsequently, section lines were tested to the north of the known mineralization along the inferred geological extension of the mineralized zone.

Method

280-ml steel containers were filled with the gases from a 4-cm diameter hole, bored to a depth of about 46 cm. It was determined that the samples could be collected immediately after establishing the sample hole.

An aliquot of the sample was transferred to an alpha scintillation cell and the cell activity was counted to a Projex Limited scintillation counter. (A rough conversion factor from cpm to picocuries per litre (pc/l) is, pc/l = 3 × cpm; Dyck, 1968.)

During these tests the sample was transferred and counted the evening of the day the samples were collected.

The early work suggested that a sample interval of 15 m was adequate to test the radioactive zones on this property.

At the time of the Rn sampling, or subsequently, scintillometer readings were taken along the Rn sampling traverses. The scintillometer readings, in the U—Th energy range, were plotted and compared with the Rn values.

Geological setting

The geology of the Faraday Township area is typical of the Grenville province. Crystalline limestones and highly metamorphosed gneisses of sedimentary and volcanic origin have apparently altered to rocks ranging from gabbro and pyroxenite to granite and pegmatite.

Of the many types of uraniferous occurrences in the area, production has been from granitic and syenitic pegmatite, or pegmatitic granite dikes. The principal-bearing minerals are uraninite and uranothorite.

On the property tested there is a north-striking, wedge-shaped limestone and gneiss area in which radioactive pegmatite dikes occur. The zone has been tested by trenching and drilling for 490 m. Some 370 m north of this area, what is believed to be the same zone have yielded interesting samples from trenches.

In the drilled area many radioactive zones were intersected, the most interesting being two shoots, 91 and 122 m long, in which individual intersections range up to 9.2 kg U_3O_8 per tonne over 0.3 m and 0.94 kg U_3O_8 per tonne over 8.7 m.

In general, averages suggest the shoots should contain from 0.8 to 0.9 kg U_3O_8 per tonne over widths of 1.5 or 1.8 m. Other similar, but discontinuous, parallel dikes occur in the zone.

Limited geological information suggests that similar conditions persist for a strike length of up to 10 km.

Results and discussion

Seven Rn soil gas traverses were made; two over the area tested by the drilling mentioned above, four on the immediate northerly extension of the zone and one about 1.5 km north of the drilling site.

The first traverse, over 122 m of grid line 33N, and shown in Fig.1, gave a very strong response over a drill-indicated section of 11.6 kg. U_3O_8 per tonne

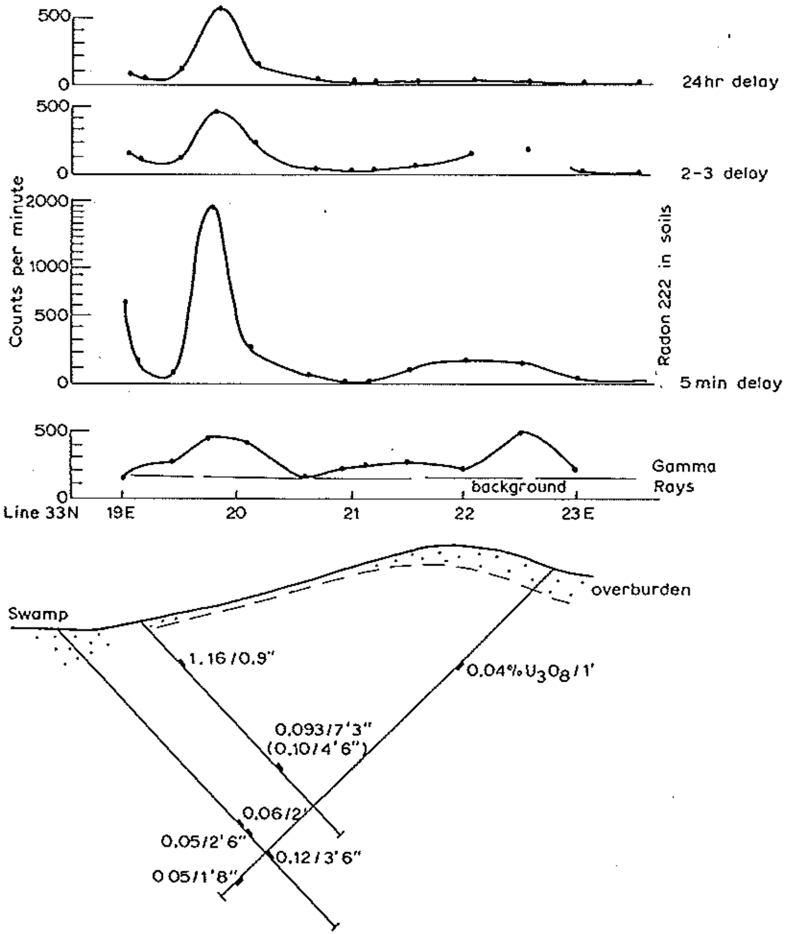


Fig.1. Radon in frozen ground. Test No.1, Bancroft, Ont., February 10, 1970. Projex Limited.

over 23 cm. The drill intersection was 12 m deep, but the mineralization probably extends to surface in an area of shallow overburden. A broad anomalous zone, well above background, appears to lie up dip from good core assays some 60 m deep. This anomaly appears to represent the near surface expression of an uraniferous zone, in which ore-grade assays occur at depth.

The second traverse as shown in Fig.2, covered 183 m over three drill holes at about grid 45N. There are three highly anomalous indications on this line. The most easterly appears to represent the up-dip extension of a zone where drill intersections were 1.13 kg U_3O_8 per tonne over 3.2 m and 4.5 kg U_3O_8 per tonne over 5.3 m, at a depth of about 91 m. Other drill holes suggest that the zone, but not the values, extend to surface.

The central Rn anomaly lies almost vertically above the U values mentioned above and may be due to secondary deposition of radioactive minerals at surface or to a source as yet unknown.

The broader westerly anomaly lies up dip of two drill-indicated uraniferous horizons.

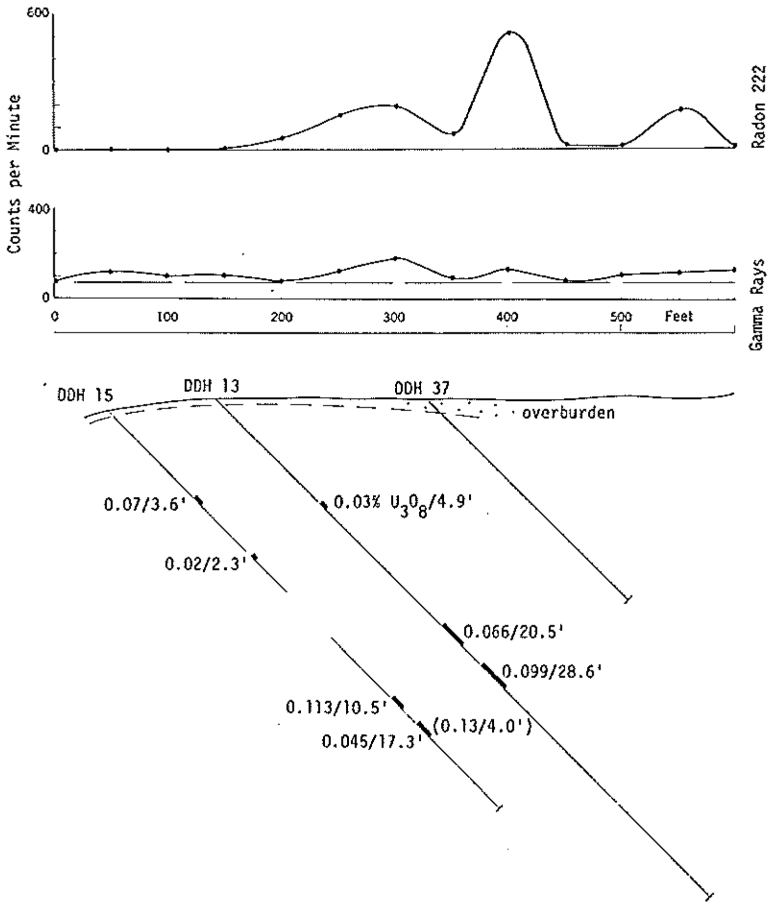


Fig.2. Radon in frozen ground. Test No.2, Bancroft, Ont., February 13, 1970. Projex Limited.

The next four Rn traverses were on grid lines 57N, 60N, 63N, and 66N, which lie 427—670 m north of the drilling at traverse No.2.

This work indicated the extensions of the two main zones intersected by the drilling, and indicated a parallel zone 122 m west, as well as two other short occurrences.

After the Rn survey, line 57N was stripped by bulldozer and trenched. Two strongly anomalous Rn values (469 cpm to 1327 cpm) were found to have been over uraniferous material from which assays were as follows: 0.5 kg U_3O_8 per tonne over 4.9 m, 1.14 kg U_3O_8 per tonne over 2.2 m and 0.77 kg U_3O_8 per tonne over 0.7 m.

91 m north of line 60 N, where an Rn count of 800 cpm was obtained, an assay of 0.3 kg U_3O_8 per tonne over 0.8 m has been reported. No other results of trenching in the area are available at this time.

The final test traverse was done along the Faraday Township north boundary across the assumed geological extension of the uraniferous zones mentioned above, and approximately 1.6 km north of the diamond drilling. Random above background values were obtained. These Rn values occur in ground with highly anomalous scintillometer readings in the U—Th energy window. It is felt that this area will prove to have a radioactive zone with a high Th/U ratio. The test did indicate continuity of the radioactive zone being tested.

Comparison with scintillometry

Rn and scintillometer anomalies were coincident, usually where overburden was thin and the source was near-surface.

Some high Rn values which could be correlated with uraniferous sources occurred without coincident anomalous scintillometry. As indicated above the sources were deep, or the overburden was heavy, or both conditions applied.

In all cases Rn backgrounds were low and anomalous values were obvious. Typically, with scintillometry the background to anomalous value ratio was high; low-grade anomalies were obscured.

RADON RETENTION BY ICE

Geological setting

The geology of the Gatineau Park has been described by Hogarth (1970). The Gatineau Hills are underlain by high-grade metamorphic (Grenville Orogeny) sequences of Precambrian gneisses and marbles, many of which are intruded by granites, syenites, peridotites and diorites. In the vicinity of Kingsmere Lake, diopside- and amphibole-rich gneisses are commonly intruded by granitic pegmatite dikes.

South of Kingsmere Lake a large calcitic marble body occurs. Some Pleistocene raised beaches and terraces can be found 1.2 km northeast and also to the east of Kingsmere Lake.

Minor occurrences of molybdenite are common throughout the Gatineau Hills, some of which are associated with traces of uraninite and uranothorite. Radioactivity is often found associated with pegmatites, particularly in the Kingsmere Lake—Fortune Lake areas.

Kingsmere Lake itself is typical of the lakes in the Gatineau Hills. It is clean and fairly shallow (~10 m) with well-defined shorelines.

Analytical method

The analytical method for Rn employed in the studies in Gatineau Park and in South March is described by Dyck (1968). Essentially, 130 ml of water is purged by bubbling air slowly (4 minutes) through the sample and letting the air pass into an evacuated cell which is coated on the inside with silver-activated ZnS. After a suitable time the sample is counted using a photomultiplier-counter assembly.

Results and discussion

On February 23, 1970, 25 water samples were collected into 45-ml plastic bottles from a hole drilled through the ice on Kingsmere Lake. The hole was some 4.5 m from the north shoreline where a small, radioactive pegmatite dike juts into the lake.

An unexpected complication arose when water was discovered between two solid layers of ice. Apparently this layer of water-saturated snow had formed during a week of mild, rainy weather early in February. A cross-section of the ice and slush layers is shown in Fig.3.

Four water samples were collected from the saturated snow layer before the second ice layer was breached. After cutting through the second ice layer 21 more samples were taken. Because no suitable sampler for taking water at depth was available, these deep-water samples were contaminated somewhat with water from the snow layer. These 21 samples were therefore divided into four groups (groups 2—5, Table I) in such a way that each group received samples sequentially as to time of sampling. In this way the contamination with water from the slush between the ice layers was essentially the same in all four groups. The treatment each group received and the Rn content of each sample are described in Table I. The general increase in Rn content of individual samples in groups 2—5 reflects the degree of mixing of deeper lake water with water from the saturated snow layer. All Rn values have been corrected for decay between time of collection and time of analysis.

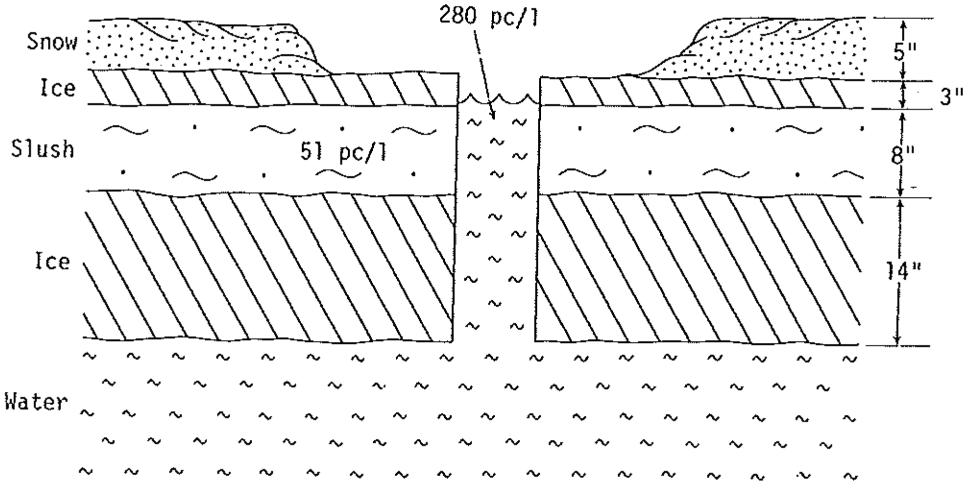


Fig.3. Cross-section of Kingsmere Lake, Gatineau Park, Quebec, 5 m from the northern shore on February 23, 1970.

Several useful conclusions can be drawn from these results. One is that although the ice acts as a cork or lid which does restrict the escape of Rn from the lake waters, it is by no means a leak-proof one — cork is perhaps an apt description. The 51-pc/l level observed in the water-saturated snow layer

TABLE I

Radon content of samples taken from an ice hole in Kingsmere Lake, Gatineau Park, Quebec, on February 23, 1970

Group	Rn content of individual samples (pc/l)					Arithmetic mean
1. Samples from water-soaked snow layer analysed immediately upon return to the laboratory	54	52	49	50	—	51
2. Mixed samples, analysed immediately upon return to the laboratory	164	162	205	275	278	217
3. Mixed samples, stored for one day at room temperature	113	120	127	183	185	146
4. Mixed samples, frozen partially outdoors for 5 hours, brought into room and analysed one day after collection	129	123	155	221;229	263	187
5. Mixed samples frozen outdoors for 21 hours, thawed in 25° C water and analysed about one day after collection.	137	167	254	242	283	217

Notes: For purposes of comparing Rn data here and in the Bancroft area a rough conversion factor which may be used is: pc/l ≈ cpm × 3 (Dyck, 1968). It is emphasized that this approximation applies only to these particular circumstances.

(slush) must have diffused through or permeated via cracks in the 35 cm lower ice layer from the Rn-enriched waters beneath. The fact that subsequently frozen samples retained Rn so well (group 5) suggests that Rn in the saturated snow layer was introduced through cracks rather than by diffusion processes. The deeper waters, prior to mixing with water above, must have contained Rn equivalent to at least 283 pc/l.

The results from group 3 samples show that Rn loss from 540 ml plastic bottles is appreciable when stored at room temperature (≈20°C) for any length of time. Group 5 results indicate clearly that the freezing of water samples immediately after collection and thawing them in room temperature tap water immediately prior to analysis, permits the retention of much more Rn. Even a partial freeze (group 4) leads to considerable improvement in Rn retention. This useful property of ice makes it practicable to conduct meaningful Rn surveys in lake waters during the winter months when samples can be conveniently preserved in this manner.

It is of some interest to compare data for winter-accumulated Rn with that found at the same sample site during the summer months. The purpose is to establish the relative degree of accumulated Rn afforded by ice cover of lake waters. Table II provides such data and indicates a winter accumulation factor of at least 10-fold, perhaps up to 20-fold.

TABLE II

Some comparative data for radon in deep lake waters during summer and winter

	Number	Range (pc/l)	Mean (pc/l)
Winter	5 (stored frozen)	137—283	217
	5 (stored unfrozen)	113—185	146
Summer	4 waters	5—15	10

Notes: (1) summer data from Dyck (1969); (2) degree of winter accumulation: at least 10-fold.

RADON IN SNOW-MELT WATERS

During the period March to May, 1974, a number of streams and low-lying areas were sampled at irregular time intervals for snow-melt waters produced by a steadily advancing spring thaw. The objective of the exercise was to investigate the abundance and variations in Rn levels in melt waters with a view to assessing the usefulness of spring thaw hydrogeochemistry of Rn as a prospecting aid in the search for U occurrences.

A measure of the magnitude of Rn accumulation beneath frozen snow and ground would also result from this work.

Two streams, one permanent (B) and the other intermittent (A) drain through a zone of known low-grade U mineralization. Both streams join downstream of the showing to form a third (D) which flows eventually to the Ottawa River.

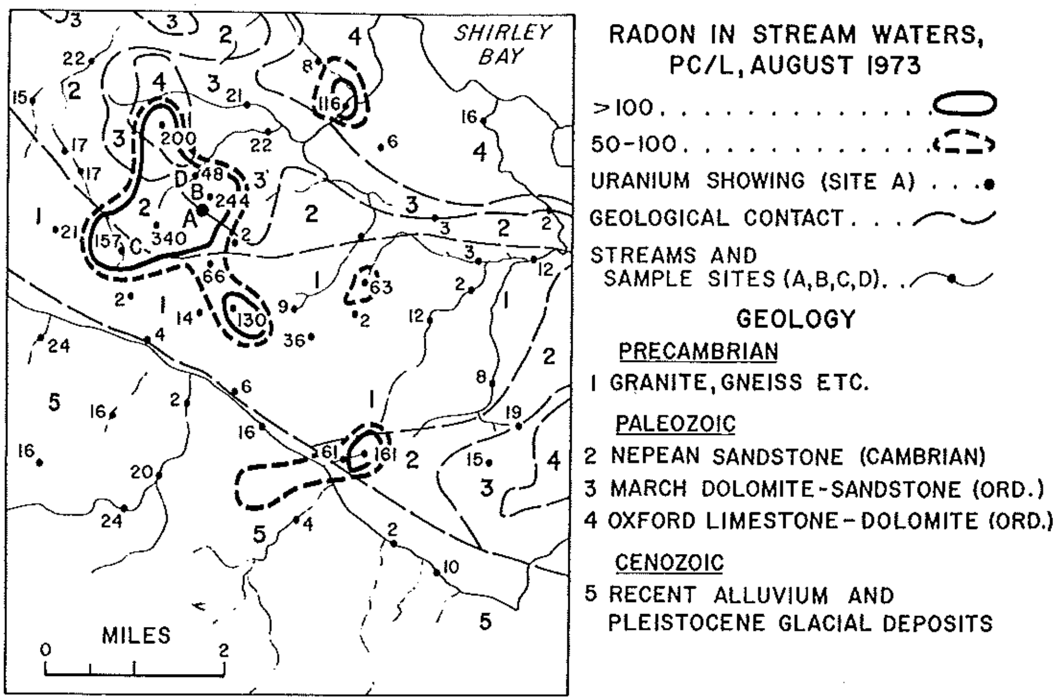
A fourth stream (C), unrelated to either the U zone or to the other three streams was also sampled. It lies some 2 km southwest of these. Relative locations of these streams and of the actual sample points are shown in Fig.4.

Rn values recorded during a regional survey of stream water in the previous summer (Jonasson and Dyck, 1974) are also indicated in Fig.4.

Geological setting

The geology of the Paleozoic sediments of the Ottawa—St. Lawrence lowlands has been described by Wilson (1946); Kirwan (1962) concentrated his studies on the Precambrian rocks of March, Huntley and Nepean Townships.

The area of interest in March Township lies on the northern flank of the sedimentary basin described by Wilson (1946). The area, typical of the whole basin, is underlain by rocks of Cambro-Ordovician age. The oldest of these, the Nepean sandstones, outcrop in the study area and are in turn conformably overlain by interbedded sandstones and dolomites (10 m) of the March Formation. Cu—U mineralization occurs sporadically in very thin, irregular conglomerate horizons near the base of the March Formation. The nature and grade of the mineralization, essentially disseminated chalcopyrite and thucholite are described in detail elsewhere (Grasty et al., 1973; Steacy et al., 1973).



GEOLOGY FROM G.S.C. MAP 414A GSC

Fig.4. Radon in stream waters (pc/l), August 1973.

The March itself is covered by dense dolomites of the Oxford Formation in the north of the study area (Fig. 4). All beds are either flat-lying or dip gently to the north (6—8°). There is little or no outcrop except in stream beds and road cuts.

Analytical method

Water samples were collected into glass bottles with care being taken to ensure *complete* filling before a crown cap seal was placed on top. In this way air bubbles into which Rn might degas were excluded. Rn was measured in the laboratory as previously described (Dyck, 1968). U was determined fluorimetrically.

Results and discussion

Data are presented in Table III for all four sample sites. Streams B, C and D are permanent, whereas stream A is in reality a low depression through which water flows only in the spring run-off. Later it is either a swamp site or else dry, as in August, 1973. Some descriptive information on the state of the thaw is also presented. Air temperatures through the first week of sampling were near 0°C, but between the 9th and 11th day after the sampling program commenced, the thaw began in earnest. By the 15th day, all streams were open and strong flows of water were observed which peaked at about the 30th day. The run-off was virtually at an end by the 49th day.

Table III includes information on both Rn and U for each water sample collected. It also reports values, from each sample site, for stream waters collected in August, 1973. Matching U (ppm) contents of stream sediments are also given.

Further information on U in soils in the immediate area of the U—Cu occurrence may be found in Charbonneau et al. (1975).

It is worth noting immediately that contrary to expectation, the Rn levels observed in spring at all sites are lower than those found the previous summer. In the case of the two sites (A and B) within the U—Cu occurrence zone, the effects of the thaw on amount of Rn are noticeable, particularly for the swamp site. At site D, some distance downstream from known U sources, the thaw effects are much weaker. In the stream (C) distant from the occurrence Rn build-up in snow-melt waters is not noticeable. Unfortunately a comparison at site A was impossible because the area dries up each summer.

Secondly, the U values in melt waters also reflect winter accumulation processes. As with Rn, U is highest near mineral sources of U, and least in the stream remote from them. Highest values of both Rn and U are found in snow-melt waters in the swampy area (A).

TABLE III

Radon (pc/l) and uranium (μg/l) in snow-melt waters

Time of thaw (days)	Thaw conditions	Site A		Site B		Site C		Site D	
		Rn	U	Rn	U	Rn	U	Rn	U
0	frozen, ice covered	10,010	14.0	162	0.3	110	0.1	179	0.8
8	frozen, ice covered	—	—	106	0.6	102	0.5	83	0.9
12	break-up	4560	19.0	165	0.1	64	0.1	177	0.6
15	break up, low flow	950	0.6	164	0.3	97	0.0	133	0.1
21	open water, high flow	5290	0.4	132	0.0	118	0.1	139	0.5
30	open water, high flow	6500	1.5	153	0.1	62	0.0	82	0.2
37	open water, low flow	6800	5.3	161	0.0	71	0.0	83	0.6
49	open water, low flow	7850	8.3	175	0.2	112	0.0	100	0.6
Summer: August 1973	generally very low water levels	dry	dry	240	0.7	150	0.1	50	2.2
U in stream sediments (ppm)		6.5		27.5		2.5		4.0	

Results indicate a trend to highest values for Rn in waters trapped beneath frozen soils, snow- or ice-covered streams. Levels for both Rn and U are depleted when stream flow is highest and rise again as the thaw ends. This is especially noticeable in stagnant waters at site A where flow was observed only between the 15th and 21st day.

For U, but not for Rn, generally higher values for waters prevail in the summer rather than in the spring; an observation which may also be due to very low water flow conditions.

The data presented illustrate a weak winter build-up of U and its daughter products, notably Rn. There was no pronounced spring flush-out of Rn and U observed in this test. A stronger manifestation of this phenomenon may be of some use in prospecting both at surface and by airborne means. As yet, the latter approach has not been attempted, although the amplification of the presence of low-grade U sources by spring thaw Rn at ground level suggests that this might be a useful approach, particularly for more deeply buried sources which may reveal their presence as escaping Rn from rising groundwaters or raised water tables. The presence of muskeg or even swamp should not deter research along these lines.

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