

Radium geochemistry applied to prospecting for uranium

By R. H. MORSE*

The author has developed a rapid method for the relative determination of Ra^{226} in sediments and has applied it to prospecting for uranium. The method discriminates against radiation from thorium, potassium and other sources.

■ Uranium²³⁸, which constitutes over 99% of natural uranium, decays through a series of daughter nuclides to lead. The important members of this series with their half-lives are arranged in order as follows:

U^{238}	4.5 billion years
U^{234}	248,000 years
Th^{230} (ionium)	80,000 years
Ra^{226} (radium)	1,600 years
Rn^{222} (radon)	3.8 days
Pb^{206}	Stable

Radon is a noble gas and is very mobile and short-lived. Because of these qualities, it has attracted interest as a geochemical indicator for uranium deposits, most recently by Dyck and Smith (1968) in their study of the Bancroft area. The immediate source of all Rn^{222} is, of course, Ra^{226} . The study of Ra^{226} has two purposes: (1) to evaluate the usefulness of radium geochemistry *per se* as a tool for uranium prospecting, and (2) to determine as much as possible the history of the Rn^{222} observed in water; that is, to determine where the transmutation $Ra^{226} \rightarrow Rn^{222}$ takes place, and hence whether Rn^{222} arrives at its present location as a gas or in the form of one of its parents.

The second goal can only be tentatively approached at this point. This paper deals mainly with the immediate application of radium geochemistry to outlining favourable areas for uranium prospecting.

Analytical method

Radium in sediments is measured by covering each sample with water and letting the daughter radon accumulate in the water. This radon is then measured with the apparatus and method developed by Willy Dyck (Dyck 1969). If the sediments are left in the water for over two weeks, Rn^{222} is nearly in equilibrium with its parent Ra^{226} , and the amount of radon in the water (in terms of radioactivity) is proportional to the amount of radium in the sediments. For shorter lengths of time, a simple correction is made: the quantity of radon measured is divided by $1 - e^{-\lambda t}$ where λ is the decay constant of Rn^{222} , 0.181 per day, and t is the time in days. To get a reasonably accurate answer, the sample must be left in the water for at least a few days.

The radium method permits analysis at any time subsequent to collection. There is no time limit during which samples must be analyzed as in the time during which it would be reasonable to analyze for radium along with copper, lead, zinc, etc., in the normal course of geochemical prospecting. An untrained worker can make twenty determinations daily. Radium in solution in water can be measured in a similar manner. In this case the

radon originally present must be swept out by bubbling or allowed to decay for two weeks or more.

The measurement of radium and radon is not affected by radiation from other sources, including potassium and the thorium decay series. The half life of Rn^{220} from the thorium series is only 54.5 seconds compared to 3.825 days for Rn^{222} . Furthermore, Rn^{220} is not followed by any long-lived alpha emitting daughters as is Rn^{222} , and the apparatus is sensitive only to alpha radiation. Samples were counted for two five minute intervals, and if significant Rn^{220} had been present the second period would have yielded a lower count than the first due to the short half-life of Rn^{220} . This did not happen. If the radioactive daughters of thorium were present, they were so much lower relative to the uranium daughters that their presence was masked by the imprecision of the method.

Although the lowest value of Ra^{226} measured was 0.022 pC/gm¹, the detection limit is about 0.01 pC/gm. The precision at the 95% confidence level is $\pm 60\%$ ².

Part of the error arises from the statistical error of low count rates. The rest of the error is probably largely due to variation in the efficiency of dissolving the radon in the water over the sample. The detection limit could be lowered and the precision improved somewhat by increasing the counting time above the fifteen minutes per sample which was used.

Sampling

Sediment samples were collected from lakes and streams over an area of 70 square miles south and west of Bancroft (see Fig. 1). This area includes three abandoned uranium mines and eight mineralized showings (Satterly 1957). Sixty-eight samples were taken from streams and twenty-seven from lakes. Some lake bottoms were sampled by dredging, the maximum depth reached being 37 feet.

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, the organic type commonly overlying the clastic type. At 38 locations both types of sediment were collected. Of the remaining locations, about half had only organic sediment and half only clastic.

¹A curie is the quantity of a radioactive nuclide in which 3.7×10^{10} disintegrations occur per second or approximately one gram of Ra^{226} . A picocurie (pC) is 10^{-12} curie.

²This is equal to

$$200 \times \sqrt{\frac{\sum_{i=1}^k \sum_{j=1}^{n_i} \left(\frac{x_{ij} - \bar{x}_i}{\bar{x}_i} \right)^2}{N - k}} \quad \%$$

where k samples were determined n_i times each; x_{ij} is the j th determination of the i th sample; \bar{x}_i is the arithmetic mean of the i th sample; and

$$N = \sum_{i=1}^k n_i$$

For a derivation of this see Cramér, p. 231-232. In this case, $N = 49$ and $k = 22$.

*Department of Geology, Queen's University

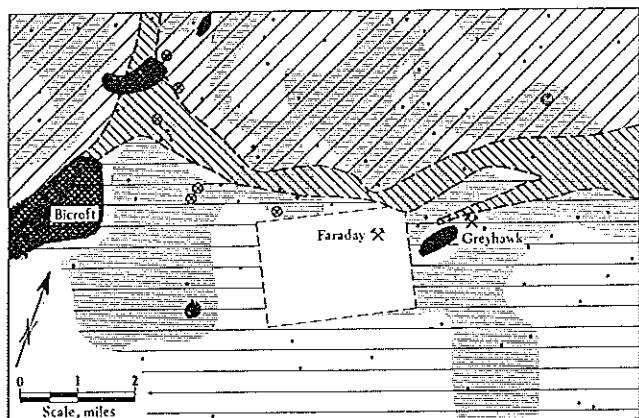


Fig. 1 Map showing distribution of radium in stream and lake sediments in the Bancroft area. Geology, generalized after Hewitt (1957), uranium mines and mineralized showings are shown for comparison. Rectangle is left blank because of the possibility that ore processing activity has added uranium and its daughters to the drainage

Results

Radium was consistently higher in organic sediments than in clastic sediments from the same location. In order to plot all the data on a single map, it is necessary first to find the equation relating the radium values in the two types of sediment, and then to express both types in the same terms. The 38 pairs mentioned above are plotted logarithmically in Fig. 2. The best least squares line is defined by

$$Y = 1.90X^{0.715}$$

where Y is the amount of radium in the organic sediment and X is the amount in the clastic sediment. The linear correlation coefficient is 0.84. If numerical values rather than logs are used, the correlation is much poorer.

For all the locations at which only clastic material was available, the radium values have been converted into "organic equivalents" using the above equation. These values together with the radium in organic sediments were contoured as shown in Fig. 1. This figure shows, in addition, all the sample points, the abandoned uranium mines,

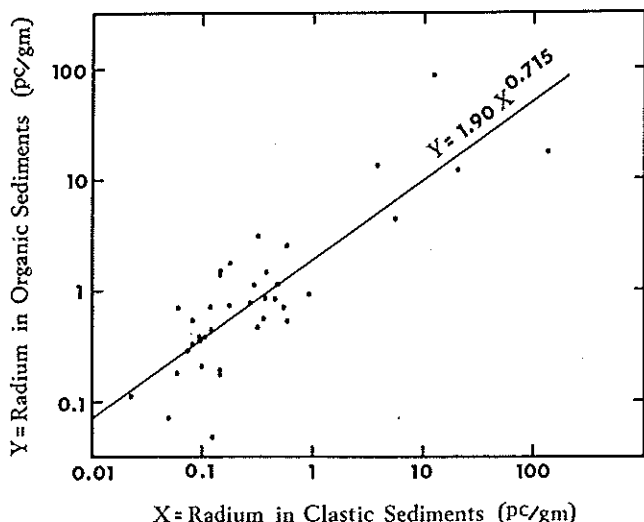


Fig. 2 Plot of radium in clastic sediments against radium in organic sediments from the same location. Best fitting line is found by the least squares method.

the mineralized showings described by Satterly (1957) and the distribution of rock types generalized after Hewitt (1957).

All the sediments downstream from the Faraday and the Bicroft mines are high in radium, but because of the possibility that ore processing activity has added uranium and its daughters to the drainage, this information has been left off the map. As a consequence, the neighbourhood of the Faraday mine was not effectively sampled, and it is shown on the map as a blank area. The high shown associated with the Greyhawk mine is downstream from the mine. This sample point is included in the figure because ore was not processed at the Greyhawk mine, but hauled away.

The correlation of high radium values with mineral rich areas is obvious in Fig. 1. In addition, both the Greyhawk and the Bicroft mines are associated with anomalous highs. A correlation is apparent also between high radium values and syenitic rocks. A similar correlation between uranium and syenitic rocks has been noted by Chamberlain (1964) and Satterly (1957).

Ra^{226} has been measured in water in the Bancroft area both by the Ontario Water Resources Commission (1968) and in the present study. Data on Ra^{226} and uranium in sediments and water, and Rn^{222} in water will eventually provide us with quantitative information on geochemical dispersion of the uranium decay series. Field, analytical, and statistical work in this direction is still in progress. However, the following conclusions are worthy of note at this point: (1) Radon in water is above the level of equilibrium with radium in water, (2) Much of the above radon comes from radium in the sediments beneath the water, and (3) At some points, radon in water is considerably above the level to be expected in equilibrium with the radium in the sediments. Such points indicate a local influx of radon-charged groundwater which has recently been in contact with uranium or its daughter products or higher radium values upstream.

Summary

Measurement of radium in sediments provides a cheap reconnaissance tool for uranium prospecting. The method discriminates between radioactivity due to uranium and that due to thorium or potassium.

Acknowledgements

The field work and part of the analytical work described here were carried out during the summer of 1968 with the support of the Geological Survey of Canada. The bulk of the analytical work was done at Queen's University using the radon system developed by Willy Dyck and loaned by the Geological Survey. Special acknowledgements are due to A. Y. Smith of the Geological Survey without whose help and encouragement the work would not have been possible, and to Prof. W. D. McCartney of Queen's University and Willy Dyck of the Geological Survey who critically read the manuscript and offered many useful suggestions during the course of the work. Mrs. Susan Carson assisted with the laboratory work. □

References

- Chamberlain, J. A., 1964. Hydrogeochemistry of uranium in the Bancroft-Haliburton region, Ontario. Geol. Surv. Can., Bull. 118.
- Gramér, Harald, 1955. The elements of probability theory. John Wiley & Sons, Inc., New York.
- Dyck, Willy, 1969. Field and laboratory methods used by the Geological Survey of Canada in geochemical surveys; No. 11. Radon determination apparatus for geochemical prospecting for uranium. Geol. Surv. Can., Paper 68-21.
- and Smith, A. Y., 1968. Use of Radon-222 in surface waters for uranium geochemical prospecting. Can. Mining J., 89, pp. 100-103.
- Hewitt, D. F., 1957. Cardiff and Faraday Townships. Ont. Dept. Mines, Map No. 1957-1.
- Ontario Water Resources Commission, 1968. Interim report on radiological water pollution in the Elliot Lake and Bancroft areas.
- Satterly, J., 1957. Radioactive mineral occurrences in the Bancroft area. Ont. Dept. Mines, 65, Pl. 6.