RADON-222, RADIUM-226, AND URANIUM IN GEORGIA PIEDMONT WELL WATER

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INTRODUCTION

Radon-222 is the gaseous daughter product of radium-226 in the uranium-238 decay chain. Since radon is an inert gas, it migrates from its location of origin in rock, soil, and saprolite into the groundwater and porespace in soil, from which it moves into the lower atmosphere. Significant amounts can enter buildings through foundations, floors, and water supplies.

High levels of radon occur in certain groundwaters (Michel, 1990). Areas of high activity were associated with rocks of granitic composition and higher-grade metamorphic lithologies in the Piedmont regions of New England, (Hall et al., 1988) and the Carolinas (Michel and Moore, 1982). In recent studies of radon in over 300 water samples from private wells in the Georgia Piedmont (Coker and Olive, 1989; Dillon, 1989; Simones et al., 1988), radon levels were found to vary widely over small areas, with some wells having extremely high levels, while others close by had moderate or low levels. An association with granitic and higher-grade metamorphic lithologies was observed in all three studies, consistent with studies from other parts of the Piedmont region.

Dillon (1989) reported radon values from 80 pCi/l to 840,000 pCi/l in 283 wells within the Inner Piedmont of Georgia. He found a log-normal distribution with a geometric mean value of 3010 pCi/l. The highest geometric mean values were associated with hydrogeologic units B (6200 pCi/l) and A (2800 pCi/l) of Radtke et al. (1986). The other units averaged 1300 pCi/l.

The purpose of this study was to test the relationship between activity levels of radon-222 and its long lived precursors, radium-226 and uranium-238, and to examine associations with lithology and common ions in the groundwater.

BACKGROUND

Radon, radium, and uranium in groundwater depend on the abundance and distribution of uranium in the original bedrock. Uranium is concentrated in the late forming minerals that commonly occur as intergranular coatings and inclusions in igneous rocks (Ragland, 1964) and in pegmatitic

areas. Sedimentary lithologies are also found to have locally high uranium concentrations. During metamorphism, uranium and other dissolved elements in hydrothermal fluids are oxidized and redistributed along foliation and bedding planes. Transport generally does not occur over long distances, thus enriched zones of uranium are close to the initial area of origin, with the uranium-bearing minerals occupying locations that will be more accessible for circulating groundwater (Lanctot et al., 1985; Gableman, 1977).

The geologic history of North Georgia is a complicated sequence of sedimentation, vulcanism, folding and metamorphic deformation. Radke et al. (1986) described the rocks of this area as predominantly metamorphic rocks with unmetamorphosed granite plutons and diabase dikes. They grouped these lithologies into water-bearing units consisting of rocks with similar character that yield water of similar chemical quality. Groundwater analyses within the study area show the water has low total dissolved solids and low ionic content. Calcium and sodium are the major cations and bicarbonate the major anion (Butler, 1990).

The units of interest within this study area are named and described as follows:

A: Amphibolite interlayered with muscovite, biotite schist and biotite gneiss, with local thin garnet quartzite.

B: Muscovite, biotite, granite gneiss, well defined layering, generally slabby; locally interlayered amphibolite and muscovite schist.

D: Biotite gneiss, well defined layering.

F: Phorphoritic biotite granite. (Elberton Granite)

I: Thinly laminated muscovite gneiss.

The bulk of the groundwater is contained in the pores and fractures of the saprolite. It slowly drains into the underlying rock, where it moves through joints, fractures, planes of schistosity, and bedding planes (Radke et al., 1986). Both the saprolite and the bedrock are considered strongly anisotropic hydrologically, with the transition zone between the bedrock and saprolite serving as a primary lateral transmitter of the groundwater into streams. Only a small portion of the groundwater enters bedrock (Velbel, 1984).

To select the wells for this study (see Fig. 1 for study area), radon data for 267 wells (Dillon, 1989; Simones, 1990) were plotted on a base map of the hydrological units of Radtke et al. (1986). Unit B in this study area is split by unit
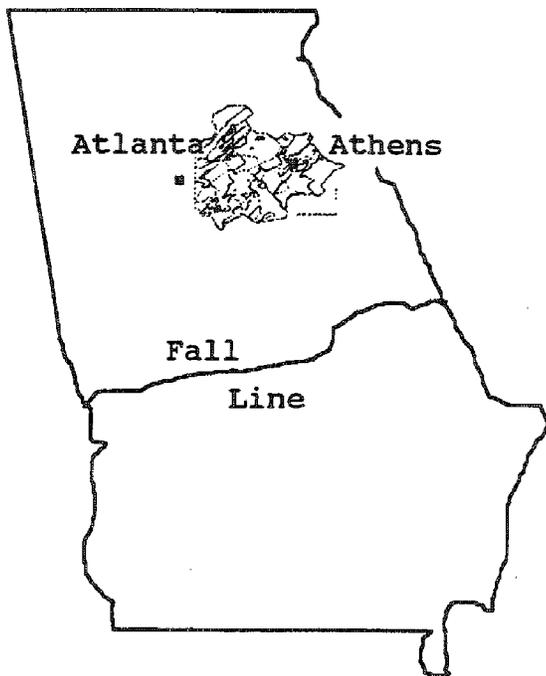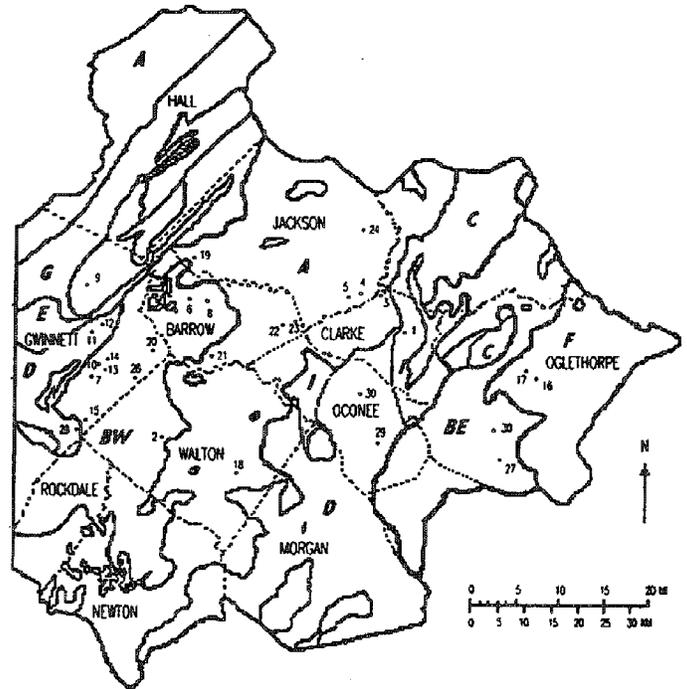
Figure 1. Location of study area.

Figure 2. Sampled wells.

A, so the western portion was renamed BW (BW coincides with mapped boundaries of the Lithonia Gneiss) and the eastern portion named BE. These three units have larger exposed areas than the other units and more wells are located in these units. The wells with high radon levels occurred most frequently near the mapped boundary of unit BW, along an E-W line across the upper center of Unit A, and in a cluster in Unit BE, near the southwestern edge of the Elberton Granite.

A set of 30 wells (Fig. 2) was selected to represent groundwater in each hydrogeologic unit for areas of high, moderate, and low radon levels. Groundwater samples from each well were analyzed for radioactive and chemical constituents.

PROCEDURES

`Wells selected for sampling were private, in moderate use, cased through saprolite and completed in bedrock (except #14). Water samples were taken at the wellhead when possible or at the closest outlet. Water was allowed to flow while pH, dissolved oxygen, conductivity, and temperature were monitored through a sampling chamber closed to the atmosphere. After these parameters stabilized, they were monitored for 20-30 minutes.

A four-liter sample was preserved with 4 ml of 16 N nitric acid for analysis of uranium and radium. Duplicate 11 ml

samples were collected with a syringe and injected into a container under a water-insoluble scintillation fluid for radon analysis. A 60 ml sample was filtered on location with a 0.45 micrometer filter and then preserved with nitric acid for cation analysis, and an unpreserved 500 ml sample was taken for anion analysis. A separate sample was placed in a sealed glass container and refrigerated for analysis of dissolved inorganic carbon. All samples were stored and analyzed according to the procedures outlined by the EPA (EPA-600/4-8-032 and 4-79-020).

Uranium was measured by coprecipitating with ferric hydroxide, eluting through an anion exchange column, and counting eluant for alpha particle activity. Radium was measured by radon emanation. A liquid scintillation counter was used to measure the activity of radon. Two samples were taken from each well and the results were averaged. Cation analysis was performed by flame atomic absorption spectophotometry. Silica concentration was obtained colorimetrically with a diode array spectrophotometer. A high pressure ion chromatograph was used for analysis of the anions. Dissolved inorganic carbon was calculated from the carbon dioxide evolved upon acidification and measured with a coulometer.

RESULTS AND DISCUSSION

Activity levels for uranium, radium-226 and radon-222 are shown in Table 1. Overall positive trends among the three

Table 1. Results of Radionuclide Analysis

| Sample No	U pCi/l	Ra-226 pCi/l	Rn-222 pCi/l	Litho-graphic Unit
1	22	2.0	14,000	I
2	38	10.0	16,000	BW
3	100	11.7	37,000	A
4	5	0.2	2,230	A
5	6	0.1	16,100	A
6	1,310	10.2	378,000	BW
7	6	0.5	28,000	BW
8	2	0.2	8,450	BW
9	577	79.4	68,900	BW
10	239	1.5	249,000	BW
11	2	1.4	5,860	D
12	1	0.1	1,110	D
13	1	1.3	12,700	BW
14	<1	0.9	6,290	BW
15	8	0.1	22,200	BW
16	3	0.6	7,990	F
17	1	0.2	8,110	F
18	<1	0.6	1,690	A
19	1	0.7	7,560	A
20	<1	<0.1	5,670	BW
21	1,930	92.3	238,000	A
22	290	1.1	125,000	A
23	53	7.3	38,600	A
24	1	0.2	1,170	A
25	62	0.3	9,630	BE
26	1	1.7	2,220	BW
27	<1	0.3	9,890	BE
28	71	<0.1	5,870	BW
29	1	0.5	874	D
30	1	0.1	250	D
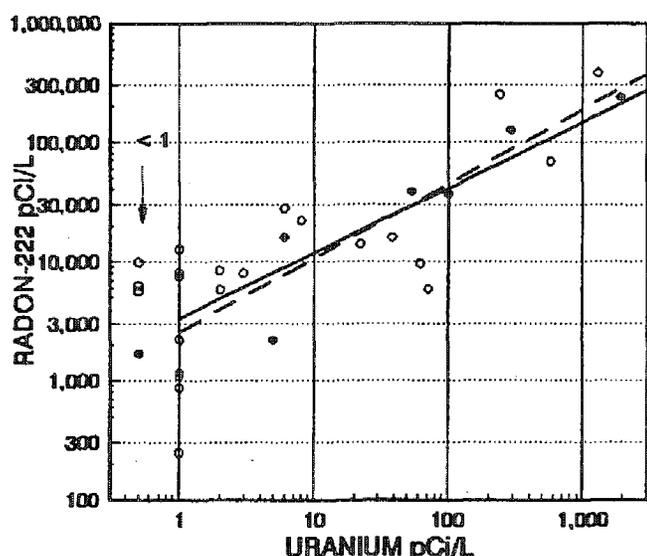
Figure 3. Radon-222 versus Uranium. (Open circles are for all wells, with $Y = 2620 \, X^{0.61}$ with $r^2 = 0.69$; solid circles are for Unit A, with $Y = 2600 \, X^{0.62}$ with $r^2 = 0.94$)

species were observed, with the strongest correlation between uranium and radon-222 [$r^2 = 0.69$ on a log/log scale (Fig. 3)]. A stronger relationship was observed when groundwater data from unit A was plotted separately.

Radon-222 enters the groundwater from 1) radium in solution, 2) radium adsorbed on fracture walls, and 3) radon emanating by alpha-particle recoil from radium in the rock itself. Because radon has the short half-life of 3.8 days, radon-222 can be directly related to radium-226 near the point of measurement.

The activity levels of radon in this study were thousands of times higher than the corresponding radium levels. Sources of radium other than that in the ground-water are the major contributor to the radon concentration.

Radium occurs in nature in a dispersed state, entering the groundwater through dissolution of uranium bearing minerals, by desorption, and by alpha-particle recoil from its immediate parent, thorium-230. Radium ions have a +2 charge, and behave similarly to Ca^{2+} and Ba^{2+}. In water of low ionic strength, sorption processes dominate for radium, and it is strongly associated with clay minerals, as it successfully competes with other alkaline earth cations for sorption sites. Feldspars, the most abundant mineral in the rock of this area, alter to form various clay minerals. Several authors have reported the existence of clay minerals on the faces of fracture surfaces, which would offer adsorption sites to chemical constituents in groundwater.

The dominant process for moving uranium-238 into the groundwater is mineral dissolution. Uranium is transported in groundwater as the uranyl ion, UO_2^{2+}, most commonly complexed by carbonate and phosphate ions or adsorbed on surfaces of ferric oxyhydroxides, clays and organic matter. Uranium does not contribute directly to the radon levels in groundwater, but high levels of uranium in water can indicate areas of higher concentrations in the rock. The correlation between uranium and radium was weak but the wells with the highest radium levels also had high uranium levels. The uranium activity is about ten times higher than the radium activity, also suggesting that most of the radium has been removed from solution.

The range of variability for the dissolved chemical species was much smaller than that for the radionuclides, with little correlation found between radioactivity levels and ionic concentrations. There was a positive correlation between pH and uranium, and a negative relationship between dissolved oxygen and uranium. In general, the best correlation between radon-222 and uranium was found for samples with the following characteristics: $pH > 6.5$, $HCO_3^- > 0.5$ meq/l, $SO_4^{2-} > 0.1$ meq/l, and $F^- > 0.05$ meq/l, with little or no PO_4^{3-} or dissolved Fe.

Uranium is known to have a tendency to be preferentially leached from fresh granitic rocks, so it could be that wells with high uranium contents are in bedrock in the early stages of decomposition. Support for this concept can be shown by examining the ratios of the concentration of uranium to

calcium and sodium, and comparing these ratios to the relative abundances of these elements in rocks that are typical of the Piedmont region. Assuming the ^{234}U activity is equal to the ^{238}U activity, 1 pCi/l is equivalent to 1.5 ug/l of uranium. All of the samples with >2 pCi/l uranium have a U/Ca ratio greater than the average ratio of these elements in granite, and from 90 to 500 times greater for the five wells where uranium is most abundant (Wampler, 1990). U/Na ratios are similarly elevated when compared to the average U/Na ratio in granite. Such water is not typical of the water draining the entire region, but if only a small fraction of water interacts with relatively fresh bedrock, then such ratios could develop. Support for this concept also comes from the observations of pH and dissolved oxygen. All the samples with >200 pCi/l uranium have a pH $\geq$ 6.5 and dissolved oxygen $\leq$ 3.2 ppm. These values suggest that the groundwater has had a chance to react with fresh bedrock in which there is iron (II) to reduce oxygen and also feldspar for hydrolysis. Wells with a pH value < 6.5 and dissolved oxygen > 6ppm all have low uranium content ($\leq$ 6 pCi/l).

SUMMARY

Since elevated radon-222 occurs in groundwater at concentrations hundreds to thousands of times higher than radium-226 (the immediate parent), radium adsorbed to rock surfaces and particulate matter and within the rock body itself must be largely the source for radon-222 in the groundwater. High levels of uranium in the groundwater do not directly contribute to the high levels of radon-222, but indicate higher than average concentrations of uranium in the rock body, which in turn will lead to high concentrations of radium-226.

The probability of finding groundwater containing elevated radionuclide levels is greater for those rock types known to have localized elevated uranium content, such as granitic gneiss and pegmatitic areas.

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