Report No. 95

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^{228}Ra, ^{226}Ra AND ^{222}Rn IN SOUTH CAROLINA GROUND WATER:

MEASUREMENT TECHNIQUES AND ISOTOPE RELATIONSHIPS

DEPARTMENT OF THE INTERIOR
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^{228}Ra, ^{226}Ra AND ^{222}Rn IN SOUTH CAROLINA GROUND WATER: MEASUREMENT TECHNIQUES AND ISOTOPE RELATIONSHIPS

by

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INTRODUCTION

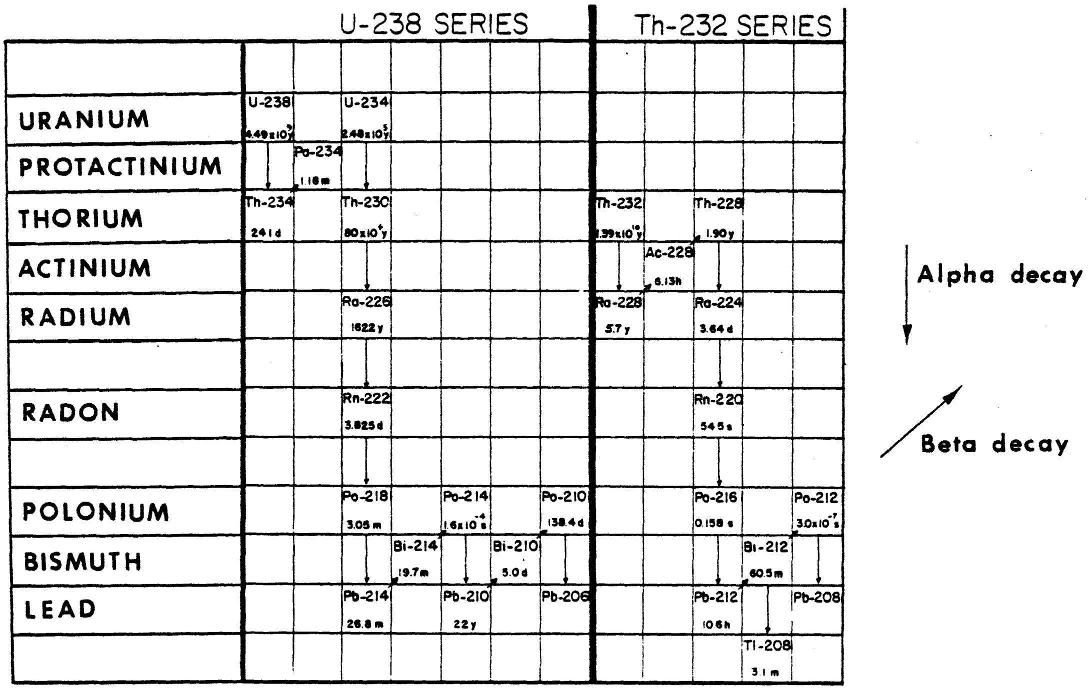
The establishment of drinking water standards for radioactivity by the Environmental Protection Agency (EPA) in 1976 initiated studies of the amounts and distribution of radioisotopes in public water supplies. Of the natural radioisotopes that occur in water, radium isotopes have the greatest potential of producing radioactive doses of consequence to man. Since radium behaves chemically much like Ca, it is a bone seeker. Once deposited in the bone, decay from radium and its daughter isotopes exposes the skeleton to high intensity alpha radiation as well as low intensity, but more penetrating, beta and gamma radiation (Figure 1).

There are two isotopes of radium which have long enough half-lives ($t_{\frac{1}{2}}$) to be of concern in drinking water (Figure 1). ^{226}Ra, an alpha emitter with a $t_{\frac{1}{2}}$ of 1622 y, is generated through decay of ^{238}U. This is the isotope commonly referred to as radium which has been measured widely. The other isotope, ^{228}Ra, is a shorter-lived, weak beta emitter ($t_{\frac{1}{2}}$ = 5.7 y) generated through decay of ^{232}Th. The interim EPA regulations state that for public water supplies:

1) The gross alpha particle activity cannot exceed 15 pCi/ℓ.
2) The combined ^{226}Ra and ^{228}Ra activity cannot exceed 5 pCi/ℓ.
3) When gross alpha particle activity exceeds 5 pCi/ℓ, the same or equivalent sample must be analyzed for ^{226}Ra.
4) When ^{226}Ra activity exceeds 3 pCi/ℓ, the same or equivalent sample must be analyzed for ^{228}Ra (EPA, 1976).

These EPA regulations assumed that ^{226}Ra would be the dominant radium isotope in public water supplies and that ^{228}Ra would occur in lower activities than ^{226}Ra in water with low radioactivity. There have been many

Figure 1. Uranium-238 and Thorium-232 decay series. Half-lives are indicated beneath the isotopes. An asterix (*) indicates an isotope of concern in this study and a plus (+) sign indicates an isotope used in the Ge(Li) detector determination of ^{228}Ra and ^{226}Ra.



reports on the activities of ^{226}Ra in ground water in varied geographic and geologic provinces (Scott and Barker 1962, Cherdyntsev 1971, Kaufmann and Bliss 1977, Michel and Moore 1980a). Radium-228 studies in ground water are few compared to ^{226}Ra due to the difficulties in present analytical procedures for ^{228}Ra. Krause (1959) reported ^{228}Ra activities in deep sandstone aquifers in Iowa, Illinois, Wisconsin and Missouri ranging from 0.9 to 7.9 pCi/ℓ, with an average of 3.30 pCi/ℓ. The ^{228}Ra/^{226}Ra activity ratio in these waters ranged from 0.04 to 2.43. Krause concluded that to calculate the total radiation dose, ^{228}Ra would have to be measured. In a recent study of Iowa public water supplies, Cochran and Hahne (1979) concluded that the level of ^{226}Ra set by EPA at which ^{228}Ra analysis is required was too high to identify all the water systems which exceed the total radium limit. Michel and Moore (1980b) found that ^{228}Ra ranged from 4.7 to 11.9 pCi/ℓ and ^{228}Ra/^{226}Ra ranged from 0.23 to 3.0 in shallow wells which were within a 4 km^2 area along the Fall Line in South Carolina. Studies of river water show that ^{228}Ra generally exceeds that of ^{226}Ra in the Amazon, Mississippi, Ganges, as well as in several minor rivers (Moore 1969, Bhat and Krishnaswami 1969). Also ^{228}Ra/^{226}Ra in low salinity coastal waters ranges generally between 1.2 and 1.8 (Li *et al.* 1979, Moore *et al.* 1980). Thus, from the assembled geochemistry data, there is no *a priori* reason to believe that ^{228}Ra will occur in natural waters in lower activities than ^{226}Ra; in fact the data suggest that the reverse may be true.

There is another problem presented by EPA regulations. The accepted technique for ^{228}Ra measurement is by extraction and beta counting of ^{228}Ac ($t_{\frac{1}{2}}$ = 6.13 hr), the immediate daughter of ^{228}Ra (Johnson 1971, EPA 1975). However, these procedures are plagued by incomplete extraction and interferences, making the analyses irreproducible by many state laboratories.

In the EPA reports on the Radium in Water Cross-Check Program, the average experimental sigma of all labs reporting ^{228}Ra in 1978 was 4.1 and in 1979 was 3.7. EPA considered a sigma greater than 3.0 to indicate serious analytical problems.

In this study, we have measured the ^{228}Ra and ^{226}Ra activities in public water supplies from a wide range of rock types in South Carolina. To do this, we developed a new technique using gamma-ray spectroscopy to measure the ^{228}Ra/^{226}Ra activity ratio. Over 180 public water supplies were analyzed by this technique. The wells sampled are located in both igneous and metasedimentary rocks in the Piedmont province and in the Coastal Plain sediments, from the Fall Line to the coast. The objectives of this study were:

1) To develop a new technique to measure accurately both ^{228}Ra and ^{226}Ra in natural water, and
2) To understand the controls on the distribution of ^{228}Ra and ^{226}Ra in different lithologies so that we can more accurately predict areas where radiological water quality may be a problem.

SAMPLING AND ANALYTICAL TECHNIQUES

Introduction

The recent introduction of high resolution, high efficiency gamma-ray detectors composed of lithium-drifted, germanium crystals has revolutionized gamma measurement techniques. These so-called Ge(Li) detectors allow the rapid measurement of relatively low-level samples without complex analytical preparation. One of the objectives of this study was the development of a new technique using Ge(Li) gamma-ray detectors for the simultaneous measurement of ^{228}Ra and ^{226}Ra in natural waters. This new technique is based on measuring the gamma-rays emitted during the α and β decays of the radium daughter isotopes. This method simplifies the analytical procedures and reduces the labor without sacrificing the precision, accuracy or lower level of detection requirements of EPA's Interium Regulations.

Field Sampling

To provide a high level of sensitivity with minimum counting times, Ra isotopes were preconcentrated in the field from large volumes of water using Mn-impregnated fibers. Moore and Reid (1973) developed a method for impregnating acrylic fiber with MnO_2 which proved effective in extracting Ra from sea water. Subsequent improvement of the MnO_2 deposition process employing permanganate oxidation of the acrylic fiber enables nearly quantitative extraction of Ra from 1000 ℓ or more of sea water (Moore 1976). The Mn-fiber removes all isotopes of Ra from sea water as well as fresh water (Moore and

Cook 1975, Michel and Moore 1980b).

In this study, commercially available 1 - 10 micron wound acrylic fiber cartridges, 25 cm in length, were used instead of loose fiber. The fiber cartridges were soaked in a 0.5 M potassium permanganate solution at 30° C for up to 7 days to ensure deposition of MnO_2 throughout the cartridge. The cartridges were then rinsed with distilled water, partially dried, and stored in plastic bags.

Each sample location was an individual public water supply well which was sampled at the well head when possible or from the water tank. A total of 182 wells in 15 different counties throughout South Carolina were sampled (Figure 2); 78 were located in the igneous and metamorphic rock aquifers of the Piedmont Province and 104 were located in the sedimentary aquifers of the Atlantic Coastal Plain Province. Wells were selected for sampling primarily at random; however some wells of known or suspected high ^{226}Ra or gross alpha particle activity were purposely sampled.

At each site water was passed through the fiber filter assembly (shown in Figure 3) at 5 - 10 ℓ/min for 1 to 4 hours to extract Ra from 100 to 1000 ℓ of water. This sample was used to measure the ^{228}Ra/^{226}Ra activity ratio of the ground water. A separate 1 to 20 ℓ sample was collected in a plastic bottle for quantitative determination of ^{226}Ra. At many sites, an additional sample was collected in 200 to 1000 ℓ evacuated glass flasks for measurement of excess ^{222}Rn. Water was allowed to overflow into a container until the container volume had been replaced three times by overflow. A hose connected to the evacuated flask was held under water and opened, allowing the water to be sucked into the flask without loss of gases. The date and time of collection was recorded so that the analysis could be corrected for ^{222}Rn decay between collection and analysis. All samples were

Figure 2. Sample location map. The symbols R and S correspond to either Piedmont crystalline rock or Coastal Plain unconsolidated sedimentary rock ground water wells respectively. The rectangular area defines the Augusta 1° x 2° topographic quadrangle.

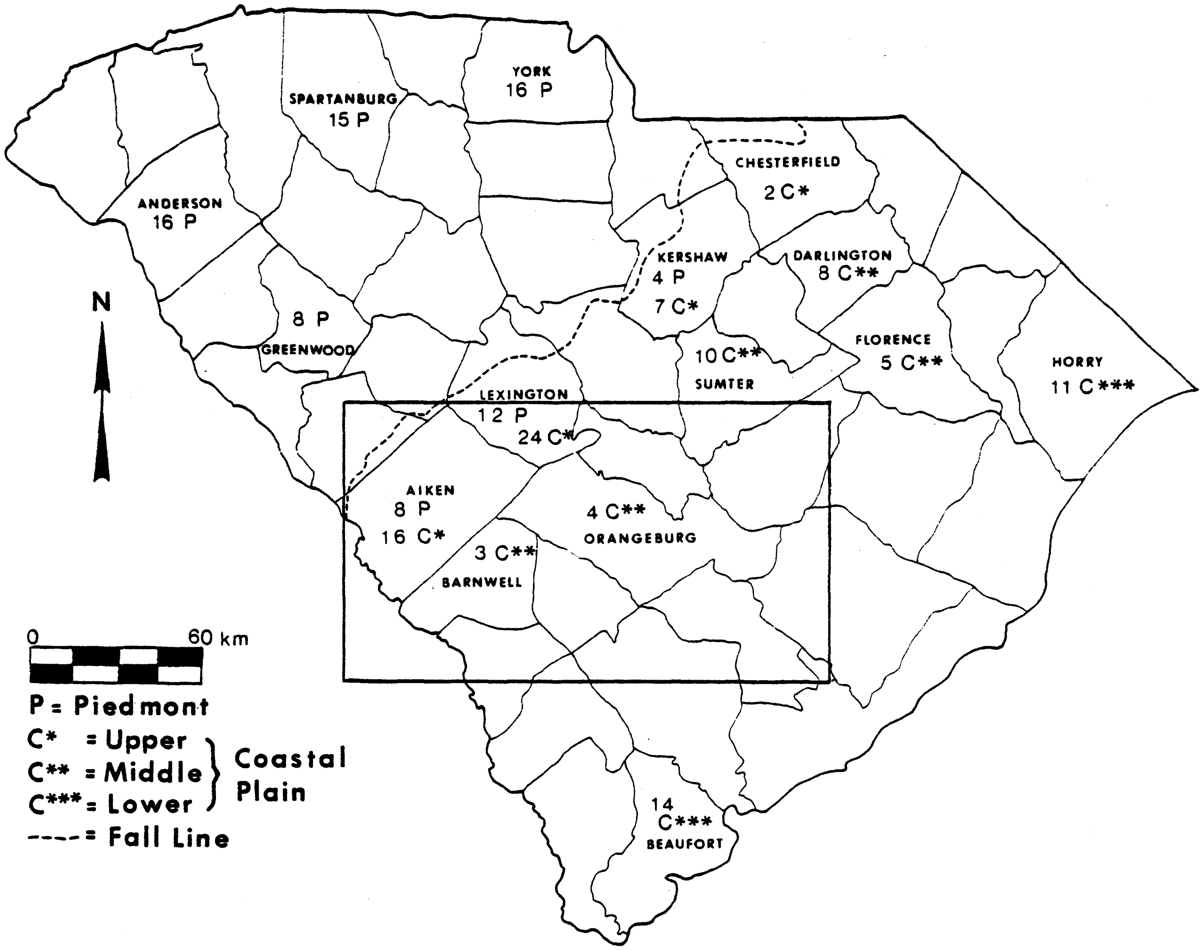
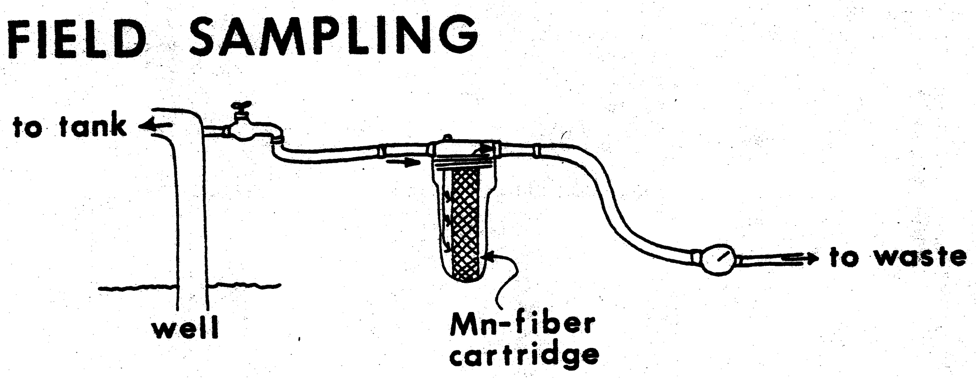


Figure 3. Field sampling system used to concentrate radium isotopes from 100 to 1000 liters of well water.



collected between April and August 1980. Michel and Moore (1980b) found little seasonal variation over a two year period in the activity of ^{226}Ra and ^{228}Ra for a single well.

Laboratory Analyses

The system used for detecting gamma-rays was a high efficiency Ge(Li) detector coupled to a preamplifier, amplifier, signal processor, and a multichannel analyzer (Figure 4). A total of 7 gamma-ray peaks, listed in Table 1, were used for calculation of the $^{228}Ra/^{226}Ra$ activity ratio. ^{226}Ra was calculated by averaging the calculated values for each of the four peaks at 295 and 352 keV for ^{214}Pb and 609 and 1120 keV for ^{214}Bi. ^{228}Ra was calculated by averaging the calculated values for each of the 338, 911 and 964 - 969 keV peaks for ^{228}Ac. These peaks were selected because of their good resolution, high percentage γ per distintegration, and high detector efficiencies. The detector efficiencies decrease exponentially with increasing energy due to the lower capture efficiency of the higher energy gamma-rays. A typical spectra is shown in Figure 4.

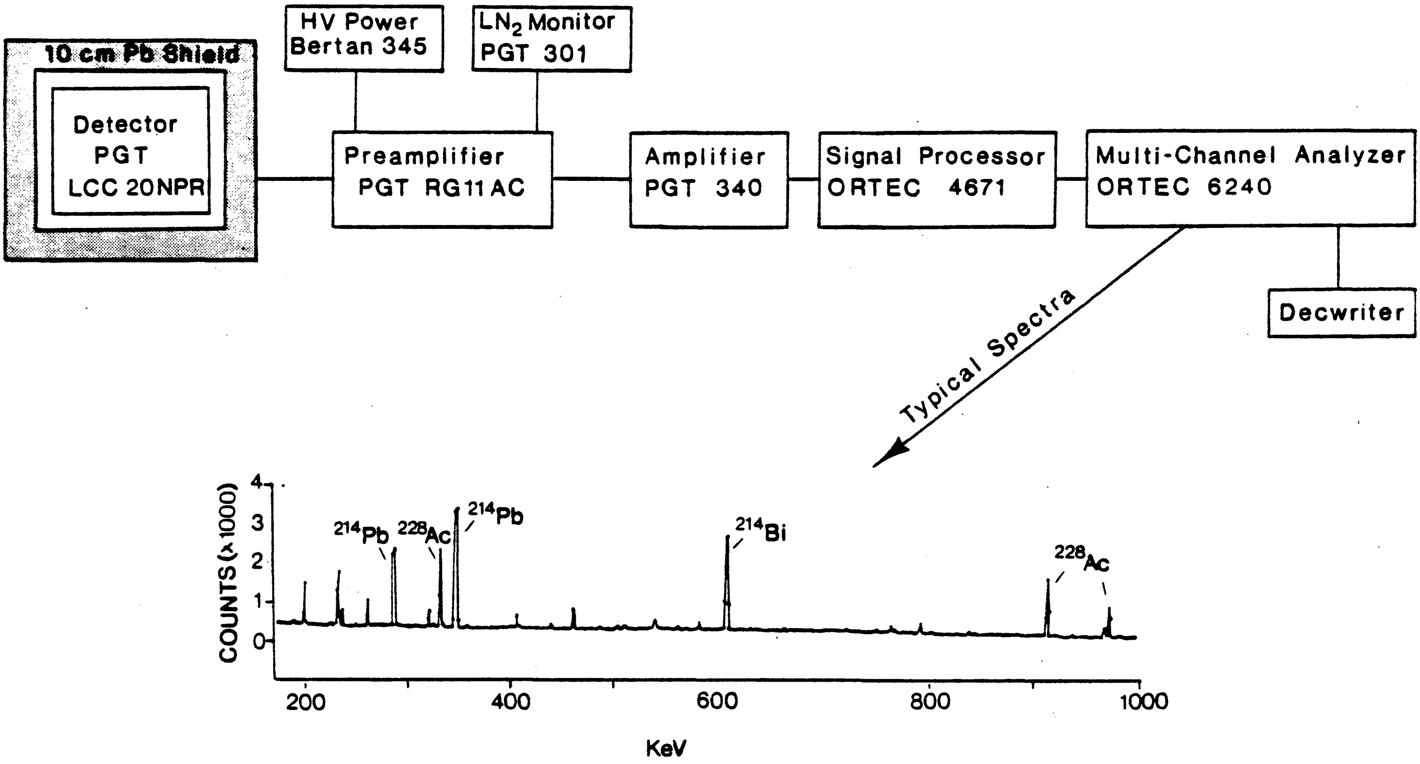
Because of the uncertainty in achieving exactly reproducable removal of isotopes from the feed water and the Mn-fiber, this method was used only to determine the $^{228}Ra/^{226}Ra$ activity ratio. A separate analysis of ^{226}Ra, which is measured easily on a small sample, was required to determine the absolute ^{228}Ra activity. The ^{226}Ra was measured using two techniques. Samples with greater than 1.0 pCi/ℓ ^{226}Ra as well as all the excess ^{222}Rn samples were analyzed using the Rn-emanation method (Mathieu 1977, Michel and Moore 1980b). To increase precision for lower activity samples, up to 20 ℓ were slowly dripped through $\sim$50 g of loose Mn-fiber to quantitatively scavenge

TABLE 1

Gamma-rays and counter efficiencies of the Ge(Li) detector used in the determination of ^{226}Ra and ^{228}Ra

Isotope	Energy (Kev)	γ/100 dis	Counter eff. (%)
	^{226}Ra		
^{214}Pb	295	17.9	8.00
^{214}Pb	352	35.0	6.87
^{214}Bi	609	43.0	3.34
^{214}Bi	1120	14.5	1.82
	^{228}Ra		
^{228}Ac	338.5	12.3	6.25
^{228}Ac	911	29.0	2.25
^{228}Ac	964-969	23.0	2.15

Figure 4. Gamma ray counting system used to measure ^{228}Ra/^{226}Ra activity ratios. A typical spectra is shown.



the Ra (Reid *et al.* 1979). The moist Mn-fiber was then placed in equilibrium flasks for analysis by Rn-emanation. Each sample was run at least twice. Experiments in our laboratory confirm that Rn emanates from the Ra adsorbed on the fiber as efficiently as from aqueous solutions. Direct analysis of the fiber saves time and prevents loss of sample during leaching.

Over 90% of the Ra on the Mn-fiber is removed by boiling the fiber in one fiber volume of 8N HCl. However, this method requires that the fiber be cut from the plastic core, generates a large amount of HCl and Cl_2 fumes, and removes Mn as well as Ra. We chose a simpler and safer technique in which the Mn-fibers were soaked overnight in cold 2N HNO_3 to desorb much of the Ra while leaving most of the MnO_2 on the fiber. In many cases, the fibers were placed in the cold nitric leach immediately after filtration in the field. Even though the cold nitric leach is not as efficient in Ra removal, over 50% of the Ra is removed using this more practical procedure.

After leaching, the cartridges were rinsed repeatedly with distilled water to make about 4 ℓ. Radium was co-precipitated with $BaSO_4$ to quantitatively scavenge the Ra by adding a solution containing 1.87 g of Ba as $BaCl_2$, then slowly adding concentrated H_2SO_4 until a precipitate formed. The $(Ra,Ba)SO_4$ precipitate was allowed to settle overnight and then was centrifuged into 20 ml scintillation vials. The samples were stored for at least 20 days to allow secular equilibrium to be reached among the daughter isotopes used for gamma-ray counting and their parent isotopes.

The Ge(Li) detector was calibrated with mixtures of various amounts of ^{226}Ra and ^{228}Ra standards which were prepared in the same manner as the samples, starting with the $BaSO_4$ precipitation. Therefore, the standards had the same matrix and geometry as the samples.

Several experiments were conducted to verify the Ge(Li) calibration and analyses. After determination of the $^{228}Ra/^{226}Ra$ activity ratio by gamma-ray spectrometry, selected standards and samples were fused with $(Na,K)_2\ CO_3$, dissolved, and split into two fractions. ^{226}Ra was determined on one fraction by the Rn-emanation technique. ^{228}Ra was determined on the other fraction by the ingrowth of its granddaughter, ^{228}Th ($t_{\frac{1}{2}}$ = 1.90 y). Thorium was removed from the solution to be measured for ^{228}Ra by repeated co-precipitations with $Fe(OH)_3$. The sample was spiked with a known activity of ^{230}Th as a chemical tracer and then stored for 10 - 60 days to allow the ^{228}Th to reach 0.98 - 5.7% equilibrium with ^{228}Ra. Th was again removed by co-precipitation with $Fe(OH)_3$, separated and purified by ion exchange, and mounted on planchets for thin source counting of ^{230}Th and ^{228}Th by alpha spectrometry.

Technique Evaluation

The absolute detector efficiencies of the gamma-ray peaks used ranged from 8.00% at 295 keV to 1.82% at 1120 keV. The calculated value of each of the individual peaks was generally within 5% of the average value. Replicate analyses of the same sample over several months gave $^{228}Ra/^{226}Ra$ activity ratios within ± 2% although the average value of each isotope varied by up to ± 10%. Any changes in the sample geometry during storage or in the location of the sample on the detector equally affect both isotopes.

Table 2 shows the results of the samples analyzed by the Ge(Li) gamma-ray technique then by the ^{228}Th-ingrowth method. When the Ge(Li) gamma-ray technique is compared with the highly accurate but more time consuming technique of allowing ^{228}Th to partially equilibrate with the ^{228}Ra in the sample, it is clear that the results are in excellent agreement. We also used this

TABLE 2

Comparison of ^{226}Ra, ^{228}Ra, and ^{228}Ra/^{226}Ra activity ratio of the same sample using two different methods

Sample	^{226}Ra (dpm)	^{228}Ra (dpm)	^{228}Ra/ ^{226}Ra[1]	Method[2]
Lx02	2129	2171	1.02	1
	2111	2202	1.04	2
Lx03	5025	4626	0.92	1
	4132	4408	1.07	2
Lx05	458	454	0.99	1
	425	468	1.10	2
Lx06	3713	5713	1.59	1
	3411	5194	1.52	2
Lx10	3081	7784	2.53	1
	2585	6294	2.43	2

[1] Values are within ±7 ^{o}/oo

[2] Method 1 - Ge(Li) spectroscopy

Method 2 - ^{226}Ra by Rn-emanation; ^{228}Ra by ^{228}Th-ingrowth technique on ground water from several wells which have been monitored for the last three years and have had constant ^{228}Ra/^{226}Ra activity ratios. The results in Table 3 show that the Ge(Li) gamma-ray analyses are consistent with earlier determinations.

During this study ground waters having a wide range of chemical compositions and levels of radioactivity were analyzed. Depending on the activity of the sample, count times ranged from 60 minutes to two days. With a two day count time, we were able to detect as low as 4 dpm bulk activity for each isotope. However, the lower detection limit is determined by the volume of water processed and the count time. Based on the results of our laboratory using the Ge(Li) technique, we estimate that ^{228}Ra/^{226}Ra activity ratios in water can be determined to within 4% given adequate sample volume. This is much better precision than procedures in routine use by laboratories reporting to the EPA Radium in Water Crosscheck Program.

TABLE 3

Radium isotope composition of ground water from wells in Leesville, S.C. using two different methods of analysis as described in the text.

Well No.	Date Collected	^{226}Ra pCi/ℓ	^{228}Ra pCi/ℓ	^{228}Ra/^{226}Ra	Method
5	Jun 78	5.6	9.7	1.73 ± 0.09	^{228}Th-Growth
	Apr 80	5.6	10.4	1.87 ± 0.15	Ge(Li)
	Jun 80	5.3	9.7	1.83 ± 0.15	"
8	Dec 77	24.2	5.5	0.23 ± 0.01	^{228}Th-Growth
	May 78	17.7	4.7	0.26 ± 0.01	"
	Aug 78	26.3	6.6	0.25 ± 0.01	"
	Oct 78	26.5	6.3	0.24 ± 0.01	"
	Apr 80	28.9	7.2	0.25 ± 0.02	Ge(Li)
	Jun 80	23.7	5.9	0.25 ± 0.02	"
14	Jun 78	6.8	9.4	1.37 ± 0.08	^{228}Th-Growth
	Oct 78	7.6	11.9	1.56 ± 0.09	"
	Apr 80	6.2	8.6	1.39 ± 0.10	Ge(Li)
	Jun 80	5.8	7.4	1.27 ± 0.10	"

RESULTS FROM SOUTH CAROLINA

Introduction

The second objective of this study was to determine the distribution of ^{228}Ra and ^{226}Ra and their relationship in a wide range of ground water types in South Carolina. Although the wells sampled were located in many different hydrogeologic settings, they can be divided into two primary groups: the rock aquifers (R) of the Piedmont Province and the sedimentary aquifers (S) of the Coastal Plain Province. Table 4 contains a complete listing of the results. Due to the fundamental differences in lithology and ground water characteristics between the Coastal Plain and Piedmont Provinces, results for ^{228}Ra, ^{226}Ra, and ^{222}Rn are addressed separately for the two regions. The distribution of these isotopes in the ground water within and between each region will be examined first using descriptive statistics and frequency histograms of the sample populations. Then the ground water of the Piedmont and Coastal Plain will be compared by the relationships between isotopes with scatter plots and regression analysis of ^{222}Rn vs ^{226}Ra and ^{228}Ra vs ^{226}Ra. By basing the comparisons between the Piedmont and Coastal Plain ground water on the interdependencies of the isotopes, as well as direct comparisons of the distributions of the isotopes, the relationship of the isotopes in the ground water to aquifer lithology can be examined.

Piedmont Crystalline Rock Aquifers

Descriptive statistics of the sample populations of ^{222}Rn, ^{226}Ra, and

Table 4

Analytical Results

LEGEND

Ak = Aiken County

An = Anderson County

Br = Barnwell County

Bf = Beaufort County

Ch = Chesterfield County

Di = Darlington County

Fl = Florence County

Gw = Greenwood County

Hy = Horry County

Ks = Kershaw County

Lx = Lexington County

Lx+ = City of Leesville, Lexington County

Mn = Monetta, Aiken County

Or = Orangeburg County

Sm = Sumter County

Sp = Spartanburg County

Yk = York County

pCi/ℓ = pico-Curie per liter

A.R. = activity ratio

R = Piedmont crystalline rock well sample

S = Coastal Plain well sample

* = Sample with total radium in excess of 5 pCi/ℓ

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}\text{Ra}}{^{226}\text{Ra}}$ A.R.	Total Ra pCi/ℓ
1	Ak	S	146	0.46	0.28	1.66	0.74
2	Ak	S	59	2.73	0.67	4.07	3.40
3	Ak	S	110	0.79	0.54	1.47	1.33
4	Ak	S	186	0.29	0.23	1.27	0.52
5	Ak	S	80	1.06	0.32	3.31	1.38
6	Ak	S	680	0.97	2.21	0.44	3.18
7	Ak	S	480	4.80	17.14	0.28	21.94*
8	Ak	S	296	0.41	0.35	1.18	0.76
9	Ak	S	198	0.87	0.68	1.28	1.55
10	Ak	S	64	2.91	0.66	4.41	3.57
11	Ak	S	---	3.26	0.77	4.24	4.03
12	Ak	S	---	1.14	0.29	3.94	1.43
13	Ak	S	---	0.96	0.58	1.65	1.54
14	Ak	S	---	1.17	0.36	3.25	1.53
15	Ak	S	---	1.27	0.54	2.35	1.81
16	Ak	S	---	0.71	0.28	2.54	0.99
17	An	R	2165	0.30	0.29	1.05	0.59
18	An	R	1099	0.40	0.30	1.32	0.70

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}Ra}{^{226}Ra}$ A.R.	Total Ra pCi/ℓ
19	An	R	1036	0.20	0.23	0.89	0.43
20	An	R	479	0.48	0.37	1.31	0.85
21	An	R	960	0.39	0.45	0.87	0.84
22	An	R	2047	0.38	0.46	0.83	0.84
23	An	R	1001	0.07	0.07	0.96	0.14
24	An	R	1607	0.33	0.16	2.06	0.49
25	An	R	2478	0.03	0.08	0.39	0.11
26	An	R	643	0.04	0.04	0.92	0.08
27	An	R	452	0.58	0.06	9.73	0.64
28	An	R	422	0.11	0.21	0.51	0.32
29	An	R	3404	0.40	0.58	0.69	0.98
30	An	R	3517	0.40	0.20	1.98	0.60
31	An	R	2224	0.96	0.42	2.29	1.38
32	An	R	---	0.49	0.17	2.89	0.66
33	Br	S	---	0.54	0.56	0.97	1.10
34	Br	S	---	0.17	1.72	0.10	1.89
35	Br	S	---	0.12	1.50	0.08	1.62
36	Bf	S	76	0.03	0.03	1.02	0.06

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}\text{Ra}}{^{226}\text{Ra}}$ A.R.	Total Ra pCi/ℓ
37	Bf	S	83	0.03	0.15	0.21	0.18
38	Bf	S	46	0.08	0.23	0.33	0.31
39	Bf	S	45	0.03	0.13	0.28	0.17
40	Bf	S	34	0.06	0.08	0.69	0.14
41	Bf	S	112	0.02	0.22	0.13	0.25
42	Bf	S	55	0.03	0.04	0.82	0.07
43	Bf	S	9	0.03	0.02	1.65	0.05
44	Bf	S	5	0.07	0.06	1.21	0.13
45	Bf	S	11	0.06	0.05	1.23	0.11
46	Bf	S	8	0.09	0.18	0.50	0.27
47	Bf	S	27	0.21	0.34	0.62	0.55
48	Bf	S	7	0.25	0.19	1.33	0.44
49	Bf	S	8	0.08	0.15	0.53	0.23
50	Bf	S	6	0.00	0.00	1.00	0.00
51	Ch	S	173	0.94	0.78	1.21	1.72
52	Ch	S	220	0.64	0.54	1.19	1.18
53	Di	S	155	1.16	0.71	1.63	1.87
54	Di	S	305	1.12	1.07	1.05	2.19

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}\text{Ra}}{^{226}\text{Ra}}$ A.R.	Total Ra pCi/ℓ
55	Di	S	218	2.70	2.23	1.21	4.93
56	Di	S	154	2.07	1.50	1.38	3.57
57	Di	S	95	0.65	0.42	1.54	1.07
58	Di	S	120	0.70	0.50	1.40	1.20
59	Di	S	23	4.12	2.19	1.88	6.31*
60	Di	S	201	6.38	2.38	2.68	8.76*
61	Fl	S	105	0.23	0.12	1.92	0.35
62	Fl	S	122	0.66	0.19	3.48	0.85
63	Fl	S	122	0.67	0.36	1.86	1.03
64	Fl	S	272	0.40	0.56	0.72	0.96
65	Fl	S	206	0.92	1.29	0.71	2.21
66	Gw	R	472	0.70	0.42	1.68	1.13
67	Gw	R	1017	0.68	0.19	3.58	0.87
68	Gw	R	2107	6.48	2.83	2.29	9.31*
69	Gw	R	1726	2.00	0.56	3.58	2.56
70	Gw	R	4648	0.03	0.17	0.16	0.20
71	Gw	R	1821	o.13	0.21	0.64	0.34
72	Gw	R	982	0.18	0.10	1.77	1.28

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}\text{Ra}}{^{226}\text{Ra}}$ A.R.	Total Ra pCi/ℓ
73	Gw	R	1365	0.28	0.07	3.97	0.35
74	Hy	S	96	0.11	0.04	2.74	0.15
75	Hy	S	97	0.05	0.02	2.35	0.07
76	Hy	S	---	0.14	0.04	3.49	0.18
77	Hy	S	105	0.06	0.03	2.06	0.09
78	Hy	S	144	0.00	0.00	2.19	0.00
79	Hy	S	126	0.00	0.00	1.75	0.00
80	Hy	S	82	0.05	0.03	1.66	0.08
81	Hy	S	108	0.03	0.02	1.74	0.05
82	Hy	S	181	0.08	0.03	2.55	0.11
83	Hy	S	115	0.06	0.02	2.86	0.08
84	Hy	S	135	0.00	0.00	2.22	0.00
85	Hy	S	105	0.16	0.07	2.32	0.23
86	Hy	S	122	0.12	0.05	2.43	0.17
87	Hy	S	139	0.12	0;06	2.08	0.18
88	Hy	S	151	0.00	0.00	0.00	0.00
89	Hy	S	113	0.00	0.00	0.00	0.00
90	Ks	S	---	0.38	0.48	0.80	0.86

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	^{228}Ra/^{226}Ra A.R.	Total Ra pCi/ℓ
91	Ks	S	252	0.00	0.00	1.02	0.00
92	Ks	S	748	1.21	1.36	0.89	2.57
93	Ks	S	488	1.73	1.80	0.96	3.53
94	Ks	S	230	1.51	2.69	0.56	4.20
95	Ks	R	3077	2.94	2.58	1.14	5.52*
96	Ks	S	607	0.25	0.11	2.30	0.36
97	Ks	R	7843	0.49	0.14	3.47	0.63
98	Ks	R	3638	0.66	0.29	2.29	0.95
99	Ks	S	101	1.40	1.15	1.22	2.55
100	Ks	S	241	4.84	4.21	1.15	9.05*
101	Ks	S	237	0.98	0.79	1.24	1.77
102	Lx	S	343	3.15	4.15	0.76	7.30*
103	Lx	S	---	1.70	1.74	0.98	3.45
104	Lx	R	2203	2.25	2.45	0.92	4.70
105	Lx	R	3764	0.61	0.44	1.38	1.05
106	Lx	R	---	0.00	0.00	1.00	0.00
107	Lx	S	---	4.53	2.85	1.59	7.38*
108	Lx	R	15288	0.43	1.17	0.37	1.60

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}\text{Ra}}{^{226}\text{Ra}}$ A.R.	Total Ra pCi/ℓ
109	Lx	S	494	3.27	1.83	1.79	5.11*
110	Lx	R	22442	1.22	2.59	0.47	3.81
111	Lx	S	265	6.81	2.69	2.53	9.50*
112	Lx	S	324	1.18	3.94	0.46	5.75*
113	Lx	S	216	0.78	1.14	0.68	1.92
114	Lx	S	235	0.68	0.64	1.06	1.32
115	Lx	S	448	1.12	1.48	0.76	2.60
116	Lx	R	4975	1.49	1.04	1.43	2.53
117	Lx	R	59430	2.43	11.57	0.21	14.00*
118	Lx	S	---	5.55	3.58	1.55	9.13*
119	Lx	R	6612	4.43	5.98	0.74	10.41*
120	Lx	S	663	4.76	8.81	0.54	13.57*
121	Lx	R	5050	1.25	1.67	0.75	2.92
122	Lx	R	7215	0.00	5.20	0.00	5.20*
123	Lx	R	9832	0.36	2.09	0.17	2.45
124	Lx	R	15322	0.10	0.22	0.44	0.32
125	Lx	S	290	6.88	4.78	1.44	11.66*
126	Lx	S	330	13.50	8.94	1.51	22.44*

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}\text{Ra}}{^{226}\text{Ra}}$ A.R.	Total Ra pCi/ℓ
127	Lx	S	---	0.22	2.79	0.08	3.01
128	Lx	R	---	1.13	11.28	0.10	12.41*
129	Mn	R	1767	0.57	0.49	1.17	1.06
130	Mn	R	3854	0.34	0.93	0.37	1.27
131	Mn	R	3126	0.31	0.50	0.62	0.81
132	Mn	R	6667	0.87	3.10	0.28	3.97
133	Mn	R	6310	1.83	1.31	1.40	3.14
134	Mn	R	5920	1.04	1.05	0.99	2.09
135	Mn	R	9210	7.25	6.71	1.08	13.96*
136	Mn	R	14800	11.94	10.47	1.14	22.41*
137	Or	S	332	1.85	9.23	0.20	11.08*
138	Or	S	142	0.08	0.15	0.54	0.23
139	Or	S	446	0.33	0.51	0.65	0.84
140	Or	S	---	0.13	0.24	0.56	0.37
141	Sm	S	164	0.85	0.42	2.02	1.27
142	Sm	S	193	0.85	0.67	1.27	1.52
143	Sm	S	108	0.65	0.34	1.92	0.99
144	Sm	S	110	0.82	0.51	1.61	1.33

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}\text{Ra}}{^{226}\text{Ra}}$ A.R.	Total Ra pCi/ℓ
145	Sm	S	299	0,94	0,62	1,52	1,56
146	Sm	S	114	0,69	0,42	1,64	1,11
147	Sm	S	162	0,00	0,00	1,21	0,00
148	Sm	S	1720	17,62	25,91	0,68	43,53*
149	Sm	S	243	0.31	2.85	0.11	3.16
150	Sm	S	107	2.34	1.71	1.37	4.05
151	Sm	S	168	1.79	1.56	1.15	3.35
152	Sp	R	882	1.60	0.47	3.40	2.07
153	Sp	R	1382	0.70	0.41	1.70	1.11
154	Sp	R	1391	0.49	0.24	2.04	0.73
155	Sp	R	1552	0.00	0.00	1,54	0.00
156	Sp	R	545	0.00	0.00	2,71	0.00
157	Sp	R	1279	0.27	0.05	5.48	0.32
158	Sp	R	1704	0.67	0.17	3,95	0.84
159	Sp	R	711	2.45	0.74	3.31	3.19
160	Sp	R	---	3,74	0.77	4.86	4.51
161	Sp	R	1348	0.88	0.21	4.17	1,09
162	Sp	R	2942	0,43	0,23	1,89	0,66

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}Ra}{^{226}Ra}$ A.R.	Total Ra pCi/ℓ
163	Sp	R	6193	0.23	0.31	0.73	0.54
164	Sp	R	3347	1.39	0.09	15.45	1.48
165	Sp	R	565	0.33	0.09	3.70	0.42
166	Sp	R	5301	0.11	0.15	0.78	0.27
167	Sp	R	---	0.19	0.09	3.27	0.38
168	Sp	R	---	0.10	0.03	3.59	0.14
169	Sp	R	---	0.00	0.00	2.01	0.00
170	Yk	R	389	0.03	0.03	1.00	0.06
171	Yk	R	7103	1.00	0.51	1.96	1.51
172	Yk	R	7951	0.84	0.69	1.22	1.53
173	Yk	R	16032	0.37	0.83	0.45	1.20
174	Yk	R	4149	0.79	0.45	1.76	1.24
175	Yk	R	---	0.65	0.26	2.49	0.91
176	Yk	R	---	0.02	0.28	0.04	0.30
177	Yk	R	---	0.03	0.03	0.95	0.06
178	Yk	R	1794	0.48	0.40	1.20	0.88
179	Yk	R	1542	0.72	0.31	2.32	1.03
180	Yk	R	---	0.24	0.08	2.99	0.32

Table 4 cont.

Sample Number	County	Aquifer Type	^{222}Rn pCi/ℓ	^{228}Ra pCi/ℓ	^{226}Ra pCi/ℓ	$\frac{^{228}Ra}{^{226}Ra}$ A.R.	Total Ra pCi/ℓ
181	Yk	R	1296	0.40	0.12	3.37	0.52
182	Yk	R	3756	0.34	0.34	1.00	0.68
183	Yk	R	555	0.53	0.66	0.81	1.19
184	Yk	R	---	1.32	1.92	0.69	3.24
185	Yk	R	---	0.89	0.77	1.15	1.66
186	Lx^{+}	S	237	9.00	7.03	1.28	16.03*
187	Lx^{+}	S	249	9.51	5.50	1.73	15.02*
188	Lx^{+}	S	1083	7.20	11.80	0.61	19.00*
189	Lx^{+}	S	1684	5.76	23.04	0.25	28.80*
190	Lx^{+}	S	---	8.13	2.71	3.00	10.84*
191	Lx^{+}	S	270	6.12	6.12	1.00	12.24*
192	Lx^{+}	R	2340	0.55	0.32	1.71	0.87
193	Lx^{+}	S	448	9.32	6.47	1.44	15.80*
194	Lx^{+}	S	287	0.00	4.58	0.00	4.58
195	Lx^{+}	S	459	0.00	3.47	0.00	3.47
196	Lx^{+}	S	164	7.68	4.34	1.77	12.02*
197	Lx^{+}	S	1212	7.66	12.98	0.59	20.64*

^{228}Ra are summarized in Table 5. Frequency histograms of the sample distributions are presented in Figure 5A. The log transformation of the values produces more compact, uniform, and continuous frequency histograms. The effect of the log transformation of the values is similar to the results observed in other radioelement studies of ground water (Scott and Barker, 1962; Kaufmann and Bliss, 1977).

The frequency histograms of log ^{228}Ra and log ^{226}Ra are both fairly evenly distributed about a central mode. The ranges of ^{228}Ra, 0.03-12 pCi/ℓ, and ^{226}Ra, 0.02-12 pCi/ℓ are similar; however, ^{228}Ra has a higher median value, 0.49 pCi/ℓ, than ^{226}Ra, 0.34 pCi/ℓ. Although ^{228}Ra is usually the dominant isotope of radium, with the median ^{228}Ra/^{226}Ra activity ratio of 1.21, the range of 0.07 to 15.45 in the activity ratios is very large. There is a distinct mode in the upper quartile of the distribution of the ^{228}Ra/^{226}Ra activity ratios in the interval from 3.32 to 406, resulting from the consistently high activity ratios for the 17 Spartanburg County samples (Table 4).

Total radium in the Piedmont rock wells is fairly uniform and has a median value of 0.89 pCi/ℓ, which is well below the 5 pCi/ℓ limit for total radium. Of the seven wells with greater than 5 pCi/ℓ in total radium, only one well, from Greenwood County, is outside the Lexington County area. It is also noteable that of the wells only one sample (5.52 pCi/ℓ) is a "borderline" case with respect to the 5 pCi/ℓ limit. The other 6 samples contain 9 or more pCi/ℓ of total radium. Why the sampling plan did not capture more of these borderline cases is not clear.

The frequency histogram of log ^{222}Rn is less variable than either of the radium isotopes, but is relatively amorphous and without dominant modes. The activities of ^{222}Rn range from above 1000 pCi/ℓ with an interquartile range of 1300-5800 pCi/ℓ and median of 2300 pCi/ℓ. The Piedmont ground water values for ^{226}Ra and ^{222}Rn are similar to values reported for the Piedmont of North Carolina

Figure 5. Frequency histograms of the log of the activities (pCi/ℓ) of ^{228}Ra, ^{226}Ra, and ^{222}Rn for Piedmont (A) and Coastal Plain (B) samples.

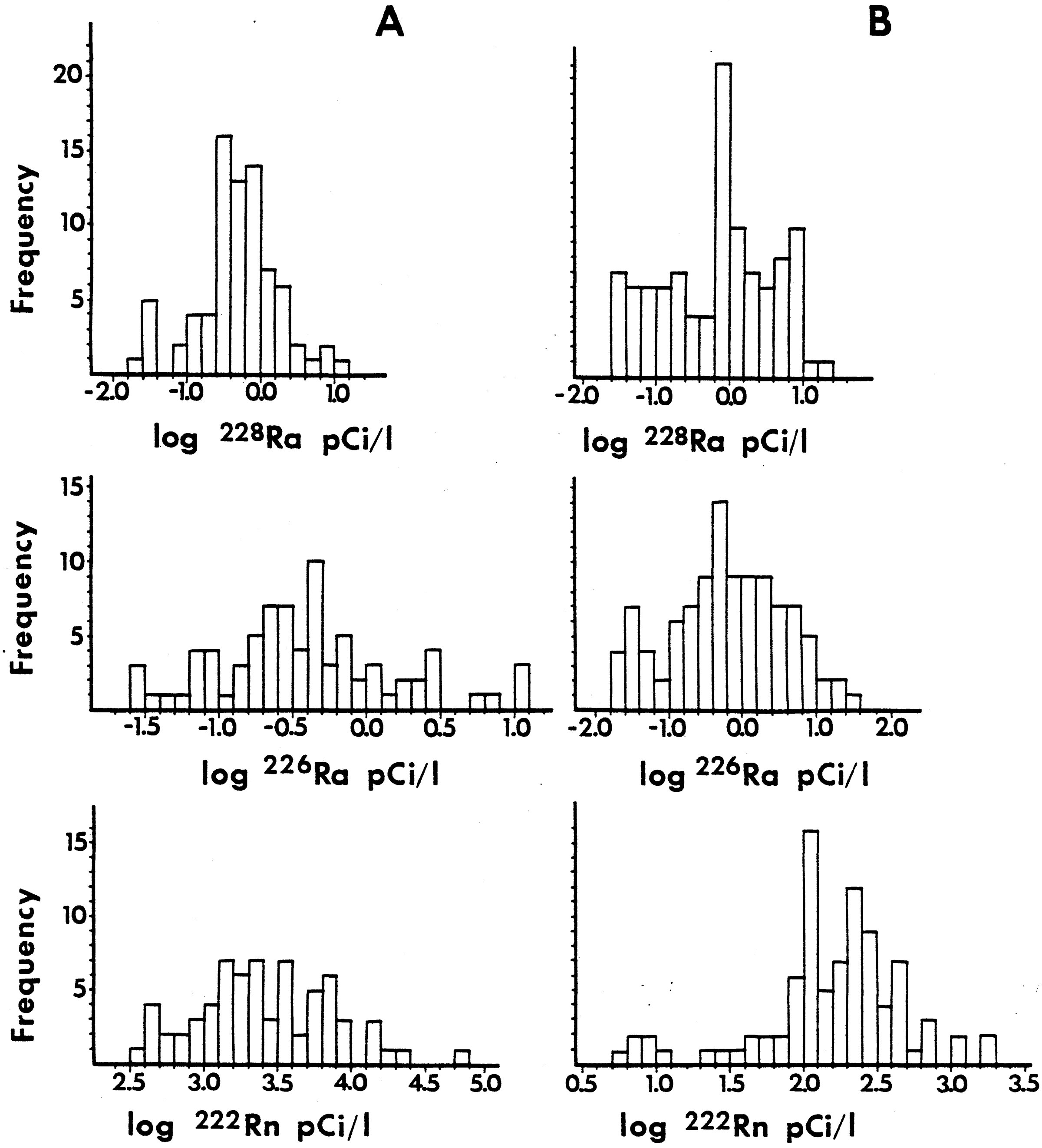


TABLE 5

Summary Statistics

	Aquifer Type	Number of Samples	Range pCi/ℓ	Interquartile Range pCi/ℓ	Median pCi/ℓ
^{226}Ra	S	104	0.02-25.91	0.20-2.22	0.60
^{226}Ra	R	78	0.03-11.57	0.17-0.78	0.35
^{228}Ra	S	104	0.03-17.62	0.16-2.60	0.83
^{228}Ra	R	78	0.02-11.94	0.30-1.01	0.49
^{228}Ra + ^{226}Ra	S	104	0.05-43.53	0.37-4.16	1.53
^{228}Ra + ^{226}Ra	R	78	0.06-22.41	0.48-2.07	0.89
^{228}Ra/^{226}Ra	S	104	0.08-4.41	0.69-1.87	1.27
^{228}Ra/^{226}Ra	R	78	0.07-15.45	0.75-2.36	1.21
^{222}Rn	S	89	5.10-1,720	103-293	168
^{222}Rn	R	68	389-59,400	1,280-5,760	2,280
^{222}Rn/^{226}Ra	S	89	10.5-6,040	83.6-515	227
^{222}Rn/^{226}Ra	R	68	744-69,600	3,150-12,700	6,060

by Lee *et al.*,(1979), but considerably lower than the values found by Asikainen and Kahlos (1979) in the crystalline rock ground water near Helsinki, Finland.

From the examination of the Piedmont sample population it appears that, in general, ^{228}Ra and ^{226}Ra are similarly distributed in ground water and rarely exceed the EPA drinking water limits. An exception to this is ground water from crystalline rocks of Lexington County area which yielded 6 of the 7 Piedmont samples exceeding the 5 pCi/ℓ limit for total radium. The highest values of ^{222}Rn were also found in Lexington County. These results indicate that these crystalline rocks contain higher concentrations of uranium and thorium than the Piedmont as a whole.

Coastal Plain Sedimentary Aquifers

A summary of the sample population descriptive statistics for the log of ^{228}Ra, ^{226}Ra, and ^{222}Rn is presented in Table 5 and frequency histograms of the sample populations are shown in Figure 5B. As for the Piedmont data, a log transformation of the Coastal Plain values results in more continuous frequency distributions.

The ranges of ^{228}Ra, 0.02 to 25.91 pCi/ℓ, and ^{226}Ra, 0.03 to 17.62 pCi/ℓ, are similar but the median value of 0.83 pCi/ℓ for ^{228}Ra is higher than the median value of 0.60 pCi/ℓ for ^{226}Ra. The frequency distributions of ^{228}Ra, ^{226}Ra, and ^{222}Rn have three modes which occur within the upper quartile, lower quartile, and the two central quartiles. Total radium values are also polymodal. Thus, it appears that the Coastal Plain samples are from three subpopulations which can also be defined geographically.

The upper quartiles of the radium distributions are dominated by samples from the extreme upper Coastal Plain and in particular, Lexington County. The lower quartiles are almost entirely composed of lower Coastal Plain samples from Beaufort and Horry Counties. There is almost no overlap in the radium values between these two geographic regions. The central quartiles contain samples from the middle Coastal Plain as well as some samples from the lower and upper Coastal Plain.

While the absolute activities of ^{228}Ra and ^{226}Ra do correspond to geographic location, the spatial distribution of the relative activities of the isotopes, defined by the ^{228}Ra/^{226}Ra activity ratios, is quite different. The distribution of the ^{228}Ra/^{226}Ra ratios has a pattern similar to the distributions of the absolute activities of the isotopes in that the distribution can be divided into three groups approximated by the quartiles. However, there is no apparent correlation between geographic location and the groupings of the radium isotope activity ratios. The upper, middle, and lower Coastal Plain samples are evenly represented in all 4 quartiles. Apparently the relationship between isotopes is fairly similar in all three geographic areas.

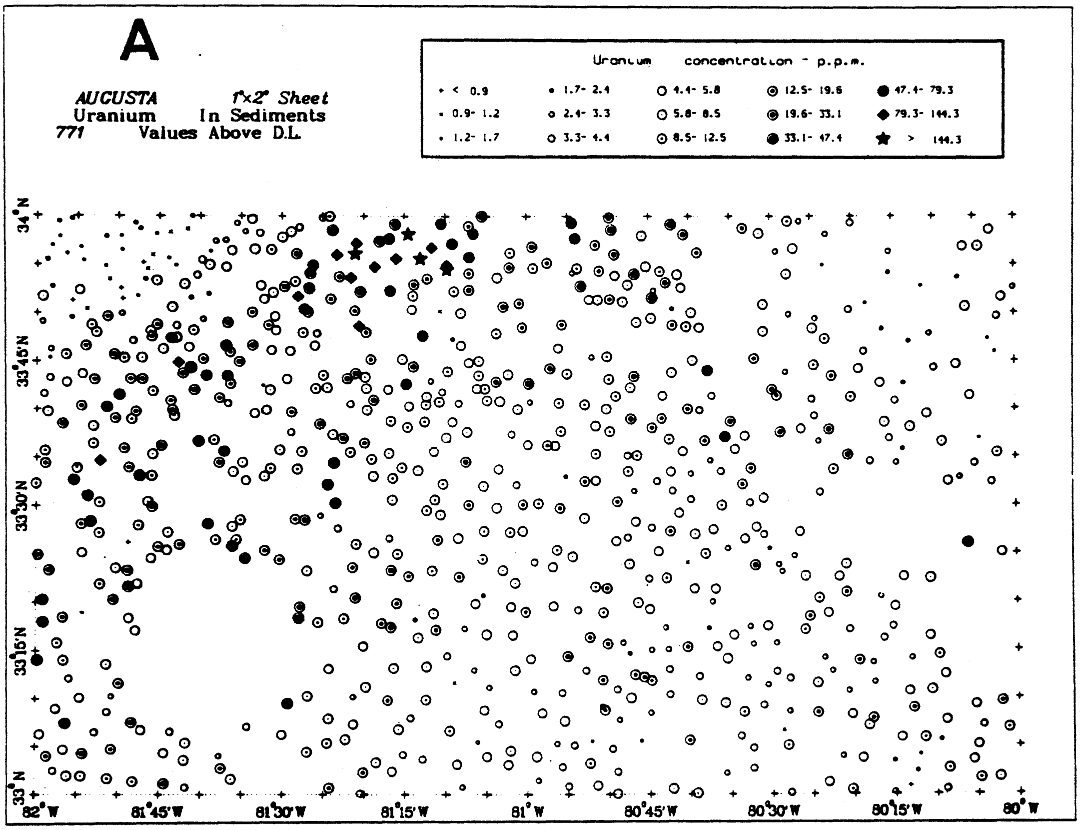
Although it appears that neither ^{228}Ra nor ^{226}Ra dominate the total radium content in any given region of the Coastal Plain, high radium activities are mostly confined to the extreme upper Coastal Plain ground waters. Of the 24 Coastal Plain samples exceeding the 5 pCi/ℓ limit for total radium, 18 are from Lexington County. The remaining samples with 5 pCi/ℓ or more of total radium are from upper Coastal Plain ground water: in Darlington, 2 samples; Aiken, 1 sample; Kershaw, 2 samples; and Sumter, 1 sample. The only other Coastal Plain sample with >5 pCi/ℓ total Ra is from Orangeburg County (the town of North) located in the middle Coastal Plain which had a total radium content of 9.23 pCi/ℓ.

The frequency distribution of the ^{222}Rn activity, shown in Figure 5B is unimodal and more uniform than the distribution of the radium isotopes. The activity ranges from 5.10 to 1,720 pCi/ℓ and has a median of 168 pCi/ℓ. ^{222}Rn is much lower in the Coastal Plain ground water than in the Piedmont. Consequently the ^{222}Rn/^{226}Ra activity ratios are also lower in the Coastal Plain ground water (Table 5). The ratios, ranging from 10.5 to 6,040 with a median of 227, are similar to those measured in the Coastal Plain ground water of North Carolina (Strain *et al.*, 1979). The highest values of ^{222}Rn are from the upper Coastal Plain and, in general, the distribution of ^{222}Rn follows the distribution of the radium isotopes; however, there is more overlap in the ^{222}Rn values between the lower and upper Coastal Plain.

The fact that the highest values of all three isotopes occur in the ground water from the unconsolidated sediments and the crystalline rocks of Lexington County suggests that the sediments overlying the crystalline rocks in this area also contain high concentrations of uranium and thorium. NURE data for the Augusta 1° x 2° quadrangle of Georgia and South Carolina (Figs. 2 & 6) reveal higher concentrations of uranium and thorium in the stream sediments of the extreme upper Coastal Plain adding support to this hypothesis.

The geographic distribution of the three isotopes indicates that the upper Coastal Plain of South Carolina is probably the area where radium isotopes pose the most serious water quality problem. The lower Coastal Plain ground water appears to contain very low activities of ^{222}Rn and radium isotopes. In fact the wells sampled in the lower Coastal Plain contain activities of ^{228}Ra and ^{226}Ra that approach the values encountered in surface water.

Figure 6. Spatial distribution of U and Th in the stream sediments of the Augusta 1° x 2° area (see Fig. 2). The plots are reproduced from Jones (1979). Note that both Th and U are higher in the upper Coastal Plain samples.


A
AUGUSTA 1°x2° Sheet
Uranium In Sediments
771 Values Above D.L.
Uranium concentration - p.p.m.
+ < 0.9
▪ 0.9- 1.2
• 1.2- 1.7
• 1.7- 2.4
o 2.4- 3.3
o 3.3- 4.4
○ 4.4- 5.8
○ 5.8- 8.5
⊙ 8.5- 12.5
◎ 12.5- 19.6
◎ 19.6- 33.1
● 33.1- 47.4
● 47.4- 79.3
◆ 79.3- 144.3
★ > 144.3
34°N
33°45'N
33°30'N
33°15'N
33°N
82°W
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81°30'W
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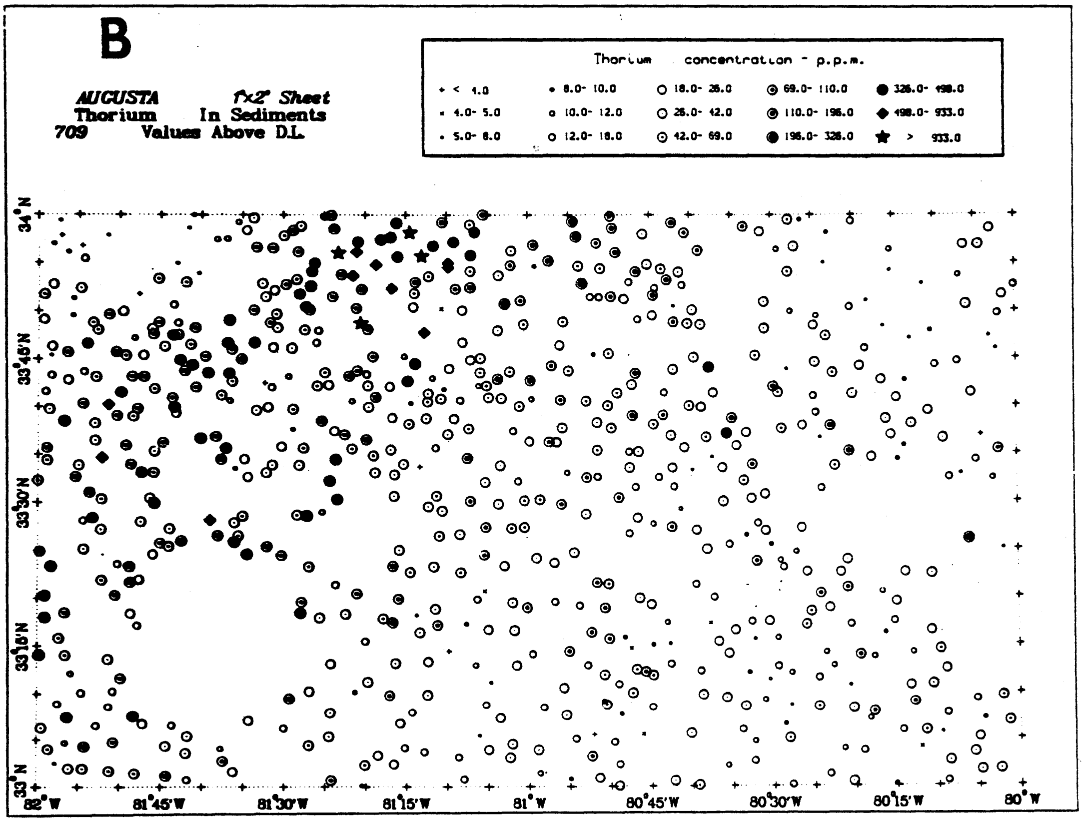
80°15'W
80°W


B
AUGUSTA 1°x2° Sheet
Thorium In Sediments
709 Values Above D.L.
Thorium concentration - p.p.m.
+ < 4.0
▪ 4.0- 5.0
• 5.0- 8.0
• 8.0- 10.0
o 10.0- 12.0
o 12.0- 18.0
○ 18.0- 26.0
○ 26.0- 42.0
⊙ 42.0- 69.0
◎ 69.0- 110.0
◎ 110.0- 196.0
● 196.0- 326.0
● 326.0- 498.0
◆ 498.0- 933.0
★ > 933.0
34°N
33°45'N
33°30'N
33°15'N
33°N
82°W
81°45'W
81°30'W
81°15'W
81°W
80°45'W
80°30'W
80°15'W
80°W

ISOTOPE CORRELATIONS

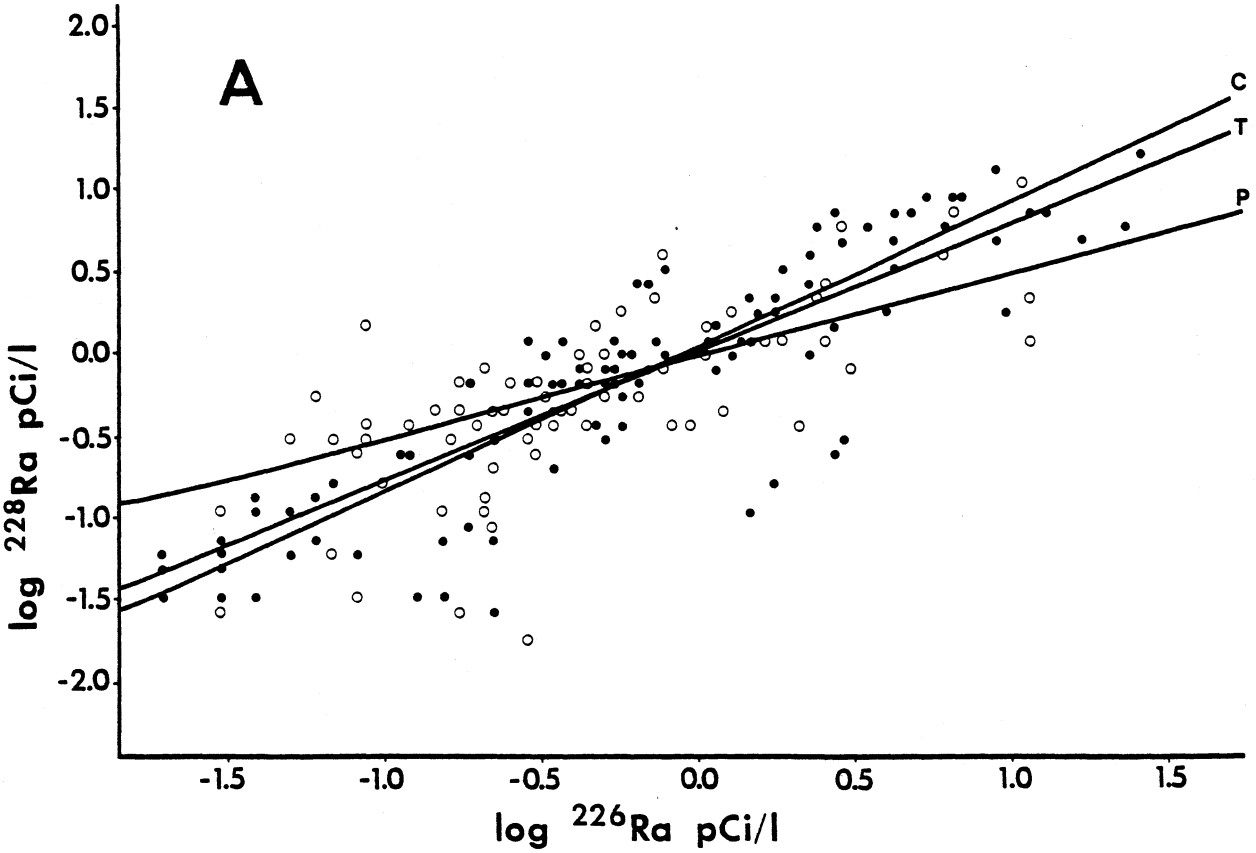
In Figure 7 scatter plots of the log ^{228}Ra and log ^{226}Ra against log ^{222}Rn are shown for both the Piedmont and Coastal Plain sample populations. The log transformation of the activities produces scatter plots to which a line can be fitted by linear least squares regression allowing comparisons between Piedmont and Coastal Plain ground water using the estimators of the best fit lines.

In Figure 7A log ^{228}Ra is plotted against log ^{226}Ra along with the best fit lines for the Piedmont and Coastal Plain sample populations. The two lines are nearly identical and can be "pooled" into a single line without a significant increase in error. Apparently there is no difference in the relationship between ^{228}Ra and ^{226}Ra in Coastal Plain and Piedmont ground water despite the lithologic differences. Similarity in the relative activities of ^{228}Ra and ^{226}Ra is also demonstrated by the ^{228}Ra/^{226}Ra activity ratios of the Piedmont and Coastal Plain ground water (Table 5). In both provinces the ground water has variable ^{228}Ra/^{226}Ra activity ratios; however, the median ratios for the Piedmont, 1.2, and Coastal Plain, 1.3, are close to estimated average crustal ^{232}Th/^{238}U activity ratios of 1.2 to 1.5, corresponding to average crustal Th/U weight ratios of 3.5 to 4.5, respectively.

It is interesting to note that, unlike the distribution of the absolute activities of ^{228}Ra and ^{226}Ra, there are no significant physiographic patterns in the ^{228}Ra/^{226}Ra activity ratios of the ground water within the Coastal Plain (Table 6). Likewise, there are no spatial patterns evident in the U/Th ratios determined for the NURE stream sediments of the Augusta 1° x 2° area (Fig. 8A). In addition, a plot of the log Th versus log U from the area

Figure 7A. Scatter plot of log ^{228}Ra against log ^{226}Ra. Open circles denote Piedmont samples (74) and solid circles denote Coastal Plain samples (108). The least squares best fit lines are also shown for Piedmont (P), Coastal Plain (C), and for total (T) sample populations. The best fit lines for the total sample population has the equation log ^{228}Ra = 0.79 ^{226}Ra + 0.00 (r = 0.83). Note that 17 obs are hidden by overlap.

7B. Scatter plot of log ^{222}Rn against log ^{226}Ra for Piedmont (68) and Coastal Plain samples (89) with the least squares best fit lines as for A. The equations for the lines are Piedmont: log ^{222}Rn = 0.5 log ^{226}Ra + 3.6 (r = 0.62); Coastal Plain: log ^{222}Rn = 0.4 log ^{226}Ra + 2.3 (4 = 0.64). Six obs are hidden by overlap.


A
C
T
P
log ^{228}Ra pCi/l
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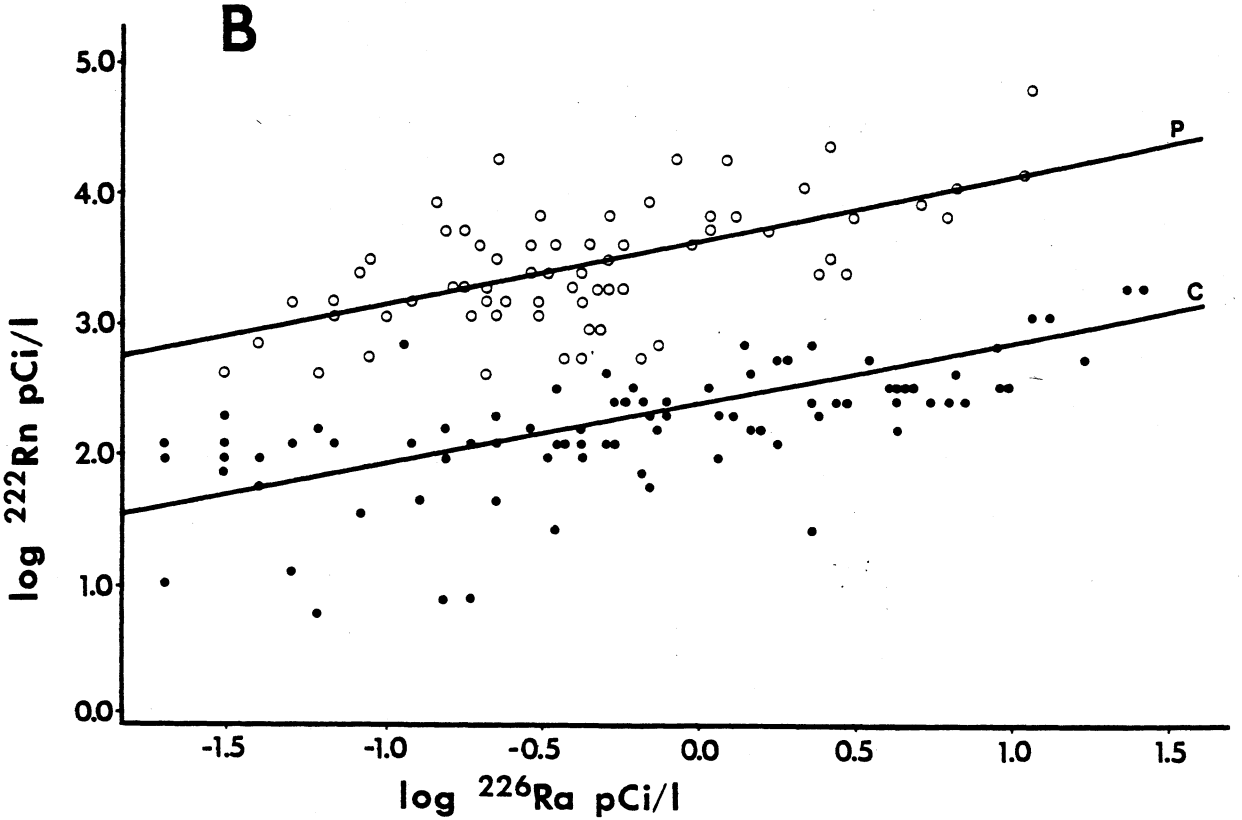
1.5
1.0
0.5
0.0
-0.5
-1.0
-1.5
-2.0
-1.5
-1.0
-0.5
0.0
0.5
1.0
1.5
log ^{226}Ra pCi/l


B
P
C
log ^{222}Rn pCi/l
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4.0
3.0
2.0
1.0
0.0
-1.5
-1.0
-0.5
0.0
0.5
1.0
1.5
log ^{226}Ra pCi/l

demonstrates that thorium and uranium are well correlated in the stream sediments (Fig. 8B).

In addition to the distribution of thorium and uranium in the aquifer solids, the relationship between ^{228}Ra and ^{226}Ra may be dependent on the age of the ground water. ^{226}Ra has a half-life 285 times greater than ^{228}Ra. If radium isotopes are removed from the aqueous phase primarily by decay mechanisms, it is expected that ground water less than a few hundred years old will have high ^{228}Ra/^{226}Ra activity ratios. For example, if ^{228}Ra and ^{226}Ra are added at a constant rate in equal activities to a given volume of water, the ^{228}Ra/^{226}Ra activity ratio at any time is given by

$$\frac{^{228}\mathrm{Ra}}{^{226}\mathrm{Ra}} = \frac{1 - e^{-\lambda_{228} t}}{1 - e^{-\lambda_{226} t}}$$

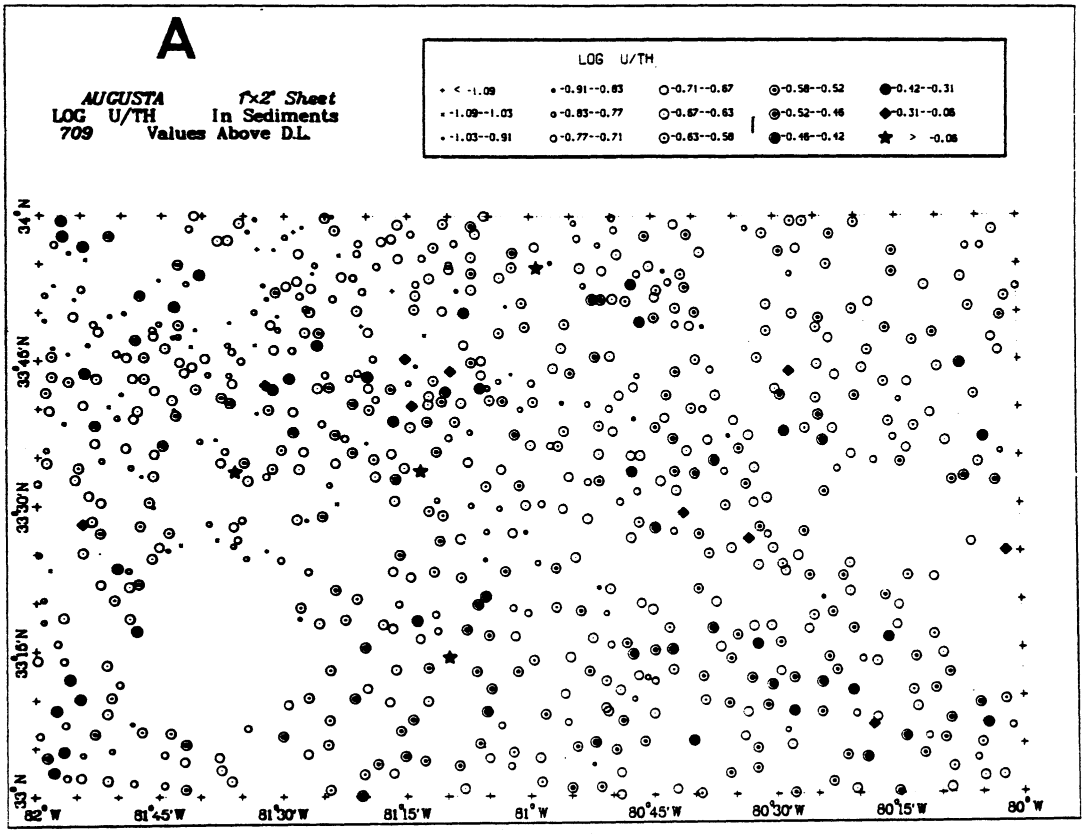
where λ_{228} and λ_{226} are the decay constants for ^{228}Ra and ^{226}Ra, respectively and t is the time. After 100 years the ^{228}Ra/^{226}Ra activity ratio is 23.9 and after 1622 years the ratio is 2. In about 5 ^{226}Ra half-lives (8,100 years) the activities of ^{228}Ra and ^{226}Ra will be equal.

The effect of ground water age on the relationship between ^{228}Ra and ^{226}Ra can be examined using the geographic distribution of the ^{228}Ra/^{226}Ra activity ratios in the ground water of the Coastal Plain. The ground water of the shallow upper Coastal Plain sediments immediately adjacent to the Fall Line are probably less than a few hundred years old whereas the ground water from the deep aquifers in the lower Coastal Plain are certainly older than 10,000 years. As was noted in the description of the sample populations, the ^{228}Ra/^{226}Ra activity ratios do not vary systematically between the upper and lower Coastal Plain ground waters. Even allowing for variations in the

Figure 8A. Statial distribution of the log of U/Th weight ratios of the Augusta 1° x 2° area stream sediments (see Fig. 2). Note the lack of any trends. Modified from Jones (1979).

8B. Scatter plot of log Th against log U for the Augusta 1° x 2° area (see Fig. 2). The letters correspond the the number of obs at each point as follows: a = 1 obs; b = 2 obs; etc. The least squares best fit line has the equation log Th = 1.14 log U + 0.11 ($r = 0.96$, $n = 709$).

D


A
AUGUSTA 1°x2° Sheet
LOG U/TH In Sediments
709 Values Above D.L.
LOG U/TH
< -1.09
-1.09--1.03
-1.03--0.91
-0.91--0.83
-0.83--0.77
-0.77--0.71
-0.71--0.67
-0.67--0.63
-0.63--0.58
-0.58--0.52
-0.52--0.46
-0.46--0.42
-0.42--0.31
-0.31--0.06
> -0.06
34°N
33°45'N
33°30'N
33°15'N
33°N
82°W
81°45'W
81°30'W
81°15'W
81°W
80°45'W
80°30'W
80°15'W
80°W

B

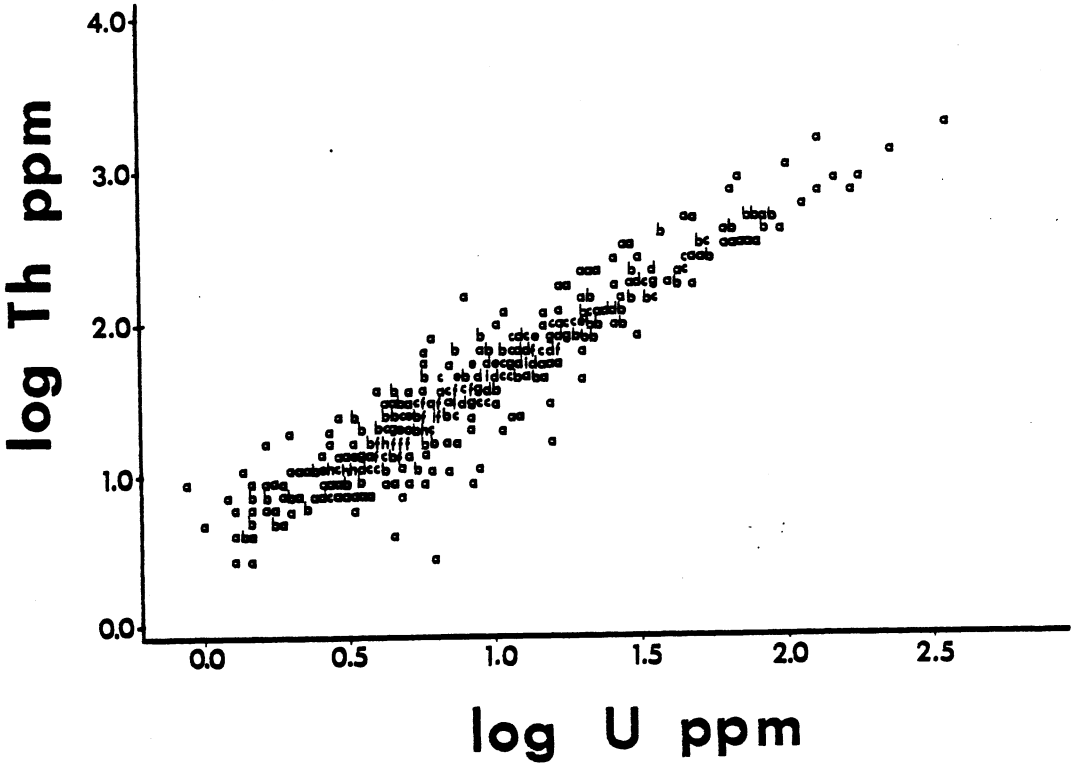

4.0
3.0
2.0
1.0
0.0
0.0
0.5
1.0
1.5
2.0
2.5
log Th ppm
log U ppm

Table 6

Summary Statistics for Subdivided Coastal Plain Samples

1-Isotopes, measured in pCi/ℓ; 2-Number of observations; 3-Subprovince*; 4-Log Mean, (Σ log x)/n; 5-Log Standard Deviation; 6-Standard Error Log Mean.

1	2	3	4	5	6
^{226}Ra	49	UCP	0.21	0.56	0.08
	30	MCP	-0.07	0.50	0.09
	25	LCP	-1.2	0.38	0.08
^{228}Ra	49	UCP	0.29	0.48	0.07
	30	MCP	-0.10	0.51	0.09
	25	LCP	-1.2	0.29	0.06
$^{228}Ra/^{226}Ra$	49	UCP	0.08	0.34	0.05
	30	MCP	-0.03	0.41	0.07
	25	LCP	0.04	0.39	0.08
^{222}Rn	39	UCP	2.5	0.33	0.05
	26	MCP	2.4	0.32	0.06
	24	LCP	1.7	0.49	0.10
$^{222}Rn/^{226}Ra$	39	UCP	2.2	0.47	0.08
	26	MCP	2.3	0.46	0.09
	24	LCP	2.9	0.73	0.15

*UCP = Upper Coastal Plain
MCP = Middle Coastal Plain
LCP = Lower Coastal Plain

relative abundances of thorium and uranium in the aquifer solids, the differences in the ^{228}Ra/^{226}Ra activity ratios in the ground water should be large due to the large differences in ground water age between the upper and lower Coastal Plain. The fact that there are no apparent differences in the ^{228}Ra/^{226}Ra activity ratios between the two regions infers that other mechanisms, such as adsorption or cation exchange, are removing radium from the aqueous phase at faster rates than decay. This hypothesis is also supported by a detailed study of the temporal and spacial variability of ^{228}Ra and ^{226}Ra in the ground water of the Leesville, S.C., plateau sediments adjacent to the Fall Line (Michel and Moore, 1980a).

In this study it was observed that while the absolute activities of ^{228}Ra and ^{226}Ra varied with time, their relative amounts remained fairly constant (see Table 3). Even though all the wells studied were located in a small area (4 km^2) and screened in similar lithologic units, each well maintained a distinct ^{228}Ra/^{226}Ra activity ratio. The implication is that ^{226}Ra and ^{228}Ra rapidly equilibrate between the aqueous and solid phases and that the solid sources of the two isotopes must be spacially similar (Michel and Moore, 1980).

If the relationship between ^{228}Ra and ^{226}Ra is independent of ground water age, then it must be predominantly dependent on the distribution of thorium and uranium in the aquifer solids. The scatter in the correlation between the two radium isotopes in the ground water implies that the relative abundances of thorium and uranium are also variable. Although the median ^{228}Ra/^{226}Ra activity ratio of the ground water is similar to the average crustal Th/U weight ratio, the scatter of the data reflects the importance of local variations in the relationship between thorium and uranium in the aquifer solids.

This hypothesis is consistent with the geochemistries of thorium and uranium. Uranium has two common oxidation states, U^{4+} and U^{6+}. In the oxidized, hexavalent state uranium can form soluble complexes; in the reduced, quadrivalent oxidation state uranium is relatively insoluble. In contrast, thorium is only stable in a quadrivalent oxidation state and does not readily form soluble complexes. Thus, uranium can be redistributed in rocks exposed to weathering processes by leaching in oxidizing environments and precipitation in reducing environments. Thorium tends to maintain its primary spacial distribution except in extremely weathered rocks (Michel *et al.*, 1978). The relationship between ^{228}Ra and ^{226}Ra in ground water may thus be a useful indicator of post-depositional behavior as well as the primary history of thorium and uranium in an aquifer. For example, a ground water from a sand aquifer with a ^{228}Ra/^{226}Ra ratio significantly less than one indicates that uranium may have been added to the aquifer solids since deposition.

The log ^{222}Rn is plotted against log ^{226}Ra in Figure 7B along with the best fit lines for the Piedmont and Coastal Plain ground water samples. In contrast to the similarities in the radium plots, the Coastal Plain and Piedmont ground water are dramatically different in terms of the relationship between ^{222}Rn and ^{226}Ra. The best fit lines for the Piedmont and Coastal Plain plots are nearly parallel with slopes of 0.5 and 0.4, respectively. However, the intercept of the Piedmont line, 3.6, is much higher than that of the Coastal Plain, 2.3. The lack of overlap between the lines is striking and there is no question that the relationships are distinct. The nearly equal slopes of the log ^{222}Rn vs log ^{226}Ra lines mean that the ^{222}Rn activity of a Piedmont ground water is usually 21 times greater than in a Coastal Plain ground water for a given ^{226}Ra activity.

The large difference in the relative activities of ^{222}Rn and ^{226}Ra between the two provinces is also reflected by the ^{222}Rn/^{226}Ra activity ratios for the Piedmont and Coastal Plain (Table 5). The median ^{222}Rn/^{226}Ra activity ratio of the Piedmont ground water is 6100 while for the Coastal Plain it is only 230. The excess ^{222}Rn content of the Piedmont ground water is similar to the findings of Lee (1979) for the North Carolina Piedmont and of Asikainen and Kahlos (1979) for the Helsinki crystalline rock ground water. The less extreme excess ^{222}Rn content of the Coastal Plain samples is similar to the results of Andrews and Lee (1979) for the ground water of the Bunter Sandstone of England and of Strain *et al.* (1979) for North Carolina Coastal Plain ground water. Although results of the subdivided Coastal Plain sample populations for the log ^{222}Rn/^{226}Ra activity ratios (Table 5) suggest that there is a significant spatial trend in the ^{222}Rn/^{226}Ra activity ratios for the Coastal Plain, the correlation is spurious. The relationship between ^{222}Rn and ^{226}Ra as measured by the ^{222}Rn/^{226}Ra activity ratio is dependent upon the absolute activity of ^{226}Ra. Since the absolute activity of ^{226}Ra is lowest in the Coastal Plain, the ^{222}Rn/^{226}Ra ratio is highest in this subprovince. The mean value of the log ^{222}Rn/^{226}Ra activity ratio for each of the sub-provinces is very close to the expected value determined by substituting the mean log ^{226}Ra value of each sub-province into the equation of the best fit line derived from the scatter plot of log ^{222}Rn against log ^{226}Ra for the whole Coastal Plain Province.

DISCUSSION

An important consideration in interpreting the ground water radioelement data is the possibility that the large differences in half lives of the isotopes result in large differences in the migration distances and hence sources of the isotopes. However, the good correlation between ^{222}Rn and ^{226}Ra, and ^{228}Ra and ^{226}Ra, in conjunction with the extreme spatial variability of the absolute activities of the isotopes within the Piedmont and Coastal Plain, suggests that the sources are in fact spatially similar. It is equally important to consider the influence of seasonal changes on the radioelement contents of the ground water. The temporal study of the Leesville ground water shows that the activities of the isotopes do not vary significantly with time. It appears then that the isotopes have spatially similar sources which are fairly stable on a monthly time scale.

The lack of regionally significant trends in the ^{228}Ra/^{226}Ra activity ratios of the ground water within and between the geologic provinces indicates that there has not been systematic redistribution of uranium relative to thorium on the scale sampled in this study. The close approximation of the average crustal ^{232}Th/^{238}U activity ratio by the median ^{228}Ra/^{226}Ra activity ratios in the Piedmont and Coastal Plain ground water demonstrates that the relative activities of the radium daughters parallel the relative abundances of thorium and uranium in the aquifer solids. Clearly, the ^{228}Ra/^{226}Ra activity ratio of ground water has potential as an exploration tool for secondary uranium mineralization. For example, a sandstone aquifer with ground water ^{228}Ra/^{226}Ra activity ratios significantly less than one may indicate the presence of secondary uranium mineralization.

While the scatter in the relationship between log ^{228}Ra and log ^{226}Ra probably reflects the importance of local variations in the relative abundances of thorium and uranium in the aquifer solids, it is more difficult to explain the overall trend in the relationship between the isotopes. The slope of the regression line determined from the log ^{228}Ra vs log ^{226}Ra plot, 0.79, means that the ^{228}Ra/^{226}Ra activity ratio decreases with increasing ^{226}Ra and ^{228}Ra contents. This infers that the Th/U ratio of the solids decreases with increasing concentrations of Th and U. However, the plot of log Th against log U (Fig. 8) for the stream sediments of the Augusta $1^{\circ} \times 2^{\circ}$ area has a slope greater than one (1.14). The contradiction between the stream sediment Th/U data and the radium ground water data may be due to a loss of leachable uranium from the stream sediments by oxidizing surface waters (Michel and Moore, 1980b). A partial loss of uranium from the stream sediments by leaching could account for the greater slope of the log Th vs log U plot than in the log ^{228}Ra vs log ^{226}Ra plot.

The extreme differences in the ^{222}Rn/^{226}Ra activity ratios between the ground water of the two geologic provinces may be related to differences in the cation sorption properties of the aquifers. The majority of aquifers sampled in the Coastal Plain were unconsolidated sands and gravels which are composed almost entirely of refractory minerals having low cation exchange capacities. The crystalline rocks of the Piedmont, being less chemically mature than the sediments of the Coastal Plain, expose more active mineral surfaces to ground water. The crystalline rock aquifers probably have a higher cation exchange capacity than the aquifers of the Coastal Plain. Radium behaves chemically like barium (Kirby and Salutsky, 1064) with an affinity for many cation exchange sites. This is shown by the Hofmeister affinity series for the alkaline earth elements $Ba^{2+} > Sr^{2+} > Ca^{2+} > Mg^{2+}$ (Stumm and Morgan, 1970). It seems reasonable that the high ^{222}Rn/^{226}Ra activity ratios of the

Piedmont ground water could be, at least in part, due to the interaction of the water with a lithology having a greater capacity to retain radium.

An interesting aspect of the relationship between ^{222}Rn and ^{226}Ra is that the $^{222}Rn/^{226}Ra$ ratio decreases with increasing ^{226}Ra and ^{222}Rn contents, implying that the farther the sampling point is from an area of enriched ^{226}Ra, the higher the $^{222}Rn/^{226}Ra$ activity ratio of the ground water. The scale length of ^{222}Rn migration by ground water transport may be greater than ^{226}Ra despite the much longer half life of ^{226}Ra. It appears that the transport of radium by ground water may be extremely limited due to continual exchange with the aquifer solids.

PREDICTION FOR THE EASTERN U.S.

The results of our investigation can be extended beyond South Carolina ground water supplies using the results of Scott and Barker (1962) on the distribution of ^{226}Ra in the ground waters of the major geographic and geologic provinces of North America. The study by Scott and Barker provides an estimate of the distribution of ^{226}Ra in the ground waters of the Atlantic and Gulf Coastal Plain as well as the entire Eastern Piedmont (see Figure 9). Our results provide an estimate of the distribution of ^{228}Ra relative to ^{226}Ra in the ground waters of these two geologic provinces. Thus, it is possible to estimate the percentage of ground water supplies which contain total radium in excess of 5 pCi/ℓ in these two regions.

This can be accomplished first by finding the probability that total radium exceeds 5 pCi/ℓ given a ^{226}Ra value. In Figure 10 this probability is plotted against ^{226}Ra. The plot has been constructed using the equation of the best fit line between log ^{228}Ra and log ^{226}Ra under the assumption that the error terms or residuals of log ^{228}Ra are independent and normally distributed. A detailed description of the calculations and assumptions involved is provided in the Appendix. After this conditional probability has been calculated it must be multiplied by the probability density function of log ^{226}Ra which is estimated from the results of Scott and Barker (1962). Plots of this function are shown in Figure 11A and B for both of the geological provinces.

The product of the conditional probability that total radium exceeds 5 pCi/ℓ and the density function of log ^{226}Ra results in the curves shown in Figure 11E and F. The total area under these curves is an estimate of the

Figure 9. This map deliniates geologic provinces samples by Scott and Barker (1962) for ground water ^{226}Ra analysis. Area I is the Atlantic and Gulf Coastal Plain and Area II is the Appalachian Piedmont.

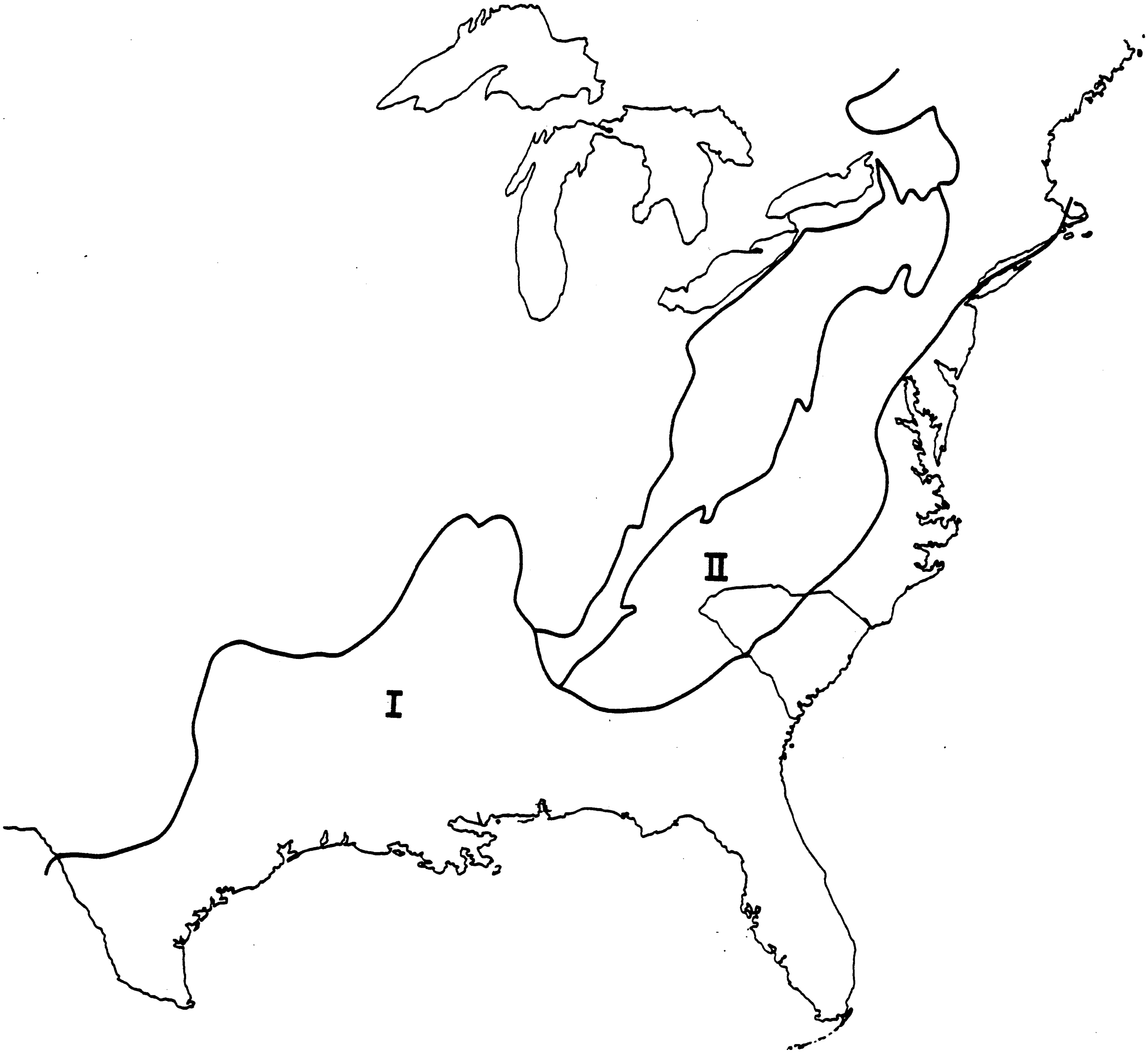


Figure 10. Plot of the probability that total radium (^{228}Ra + ^{226}Ra) exceeds 5 pCi/ℓ against ^{226}Ra in pCi/ℓ. (See Appendix for details). Note the lack of large changes in slope indicating that the selection of a suitable screening level of ^{226}Ra for ^{228}Ra analysis will be difficult for South Carolina ground water supplies.

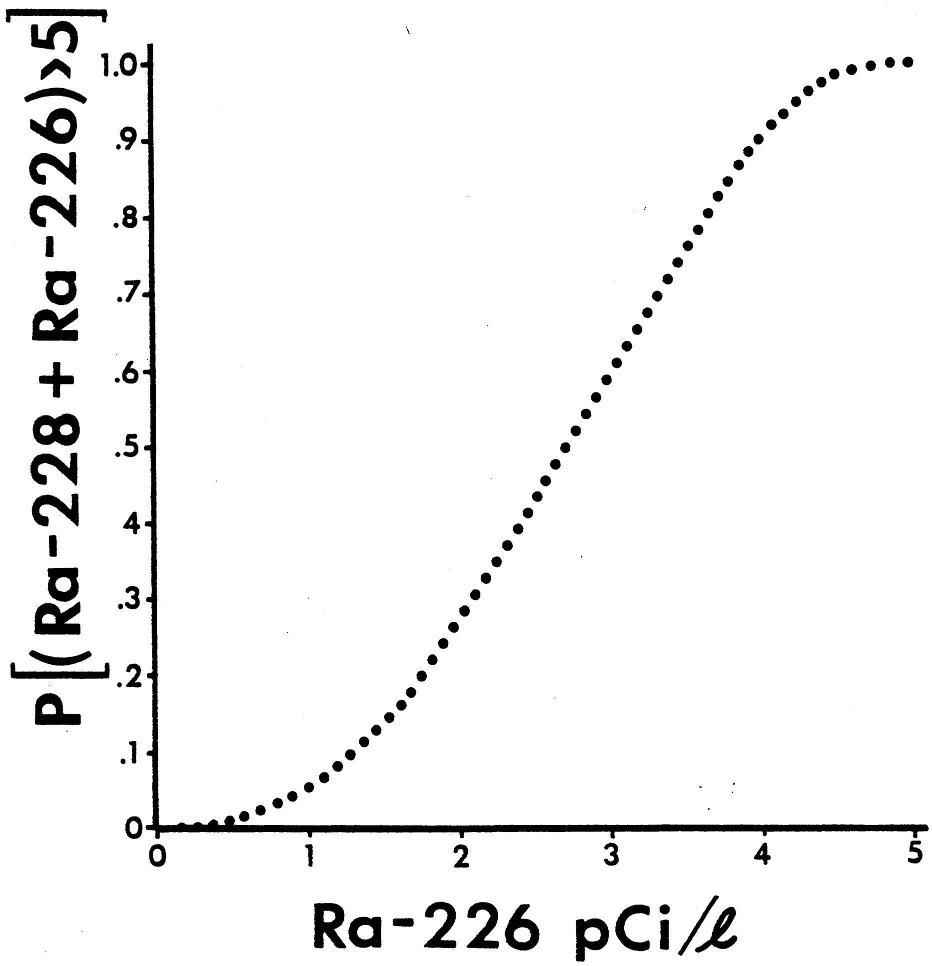


Figure 11A & B. Plots of the estimated probability density function of log ^{226}Ra for Appalachian Orogenic Belt (Fig. A) and Atlantic and Gulf Coastal Plain (Fig. B) ground water. The plots were constructed with data from a study by Scott and Barker (1962) under the assumption that the distribution of log ^{226}Ra can be approximated by a normal distribution denoted by N (μ, σ) where μ is the mean and σ is the standard deviation. Appalachian ground water has a log ^{226}Ra distribution of $\sim$ N (-1.00, 0.67) and Coastal Plain ground water has a log ^{226}Ra distribution of $\sim$ N (-0.70, 0.64) shown in Figure 11A and B respectively.

Figure 11C. Frequency histogram of the residuals of log ^{228}Ra determined from the best fit line of Figure 7A. The dotted line corresponds to the normal distribution with a mean (0.00) and standard deviation (0.85) estimated from the sample population.

Figure 11D. Scatter plot of the residuals of log ^{228}Ra against log ^{226}Ra. Open circles denote Piedmont and solid circles denote Coastal Plain samples. The residuals are from the overall best fit relationship between log ^{228}Ra and log ^{226}Ra (see Fig. 7A). Note the independence of the residuals. 17 obs are hidden by overlap.

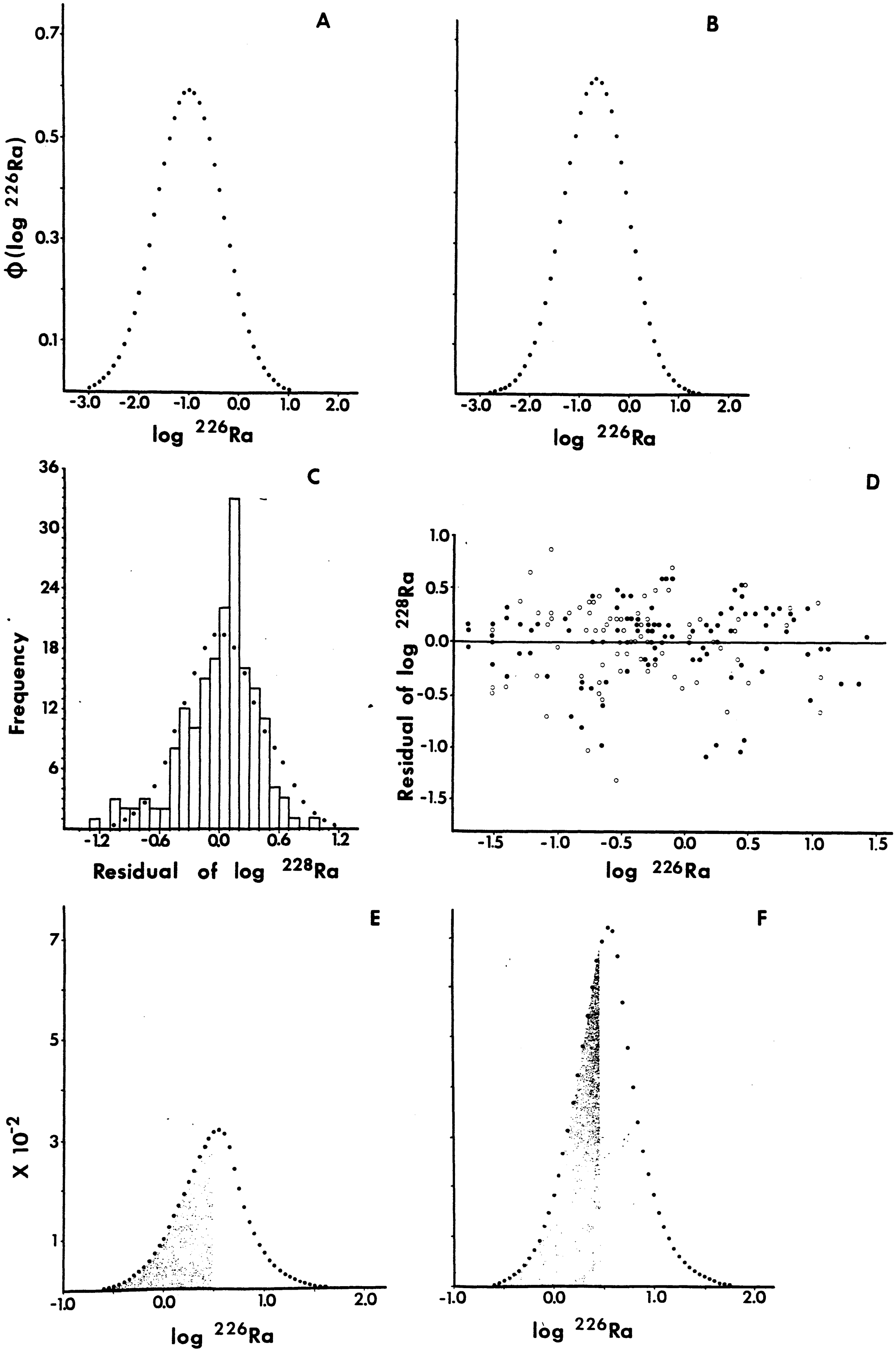
Figure 11E. Plot of the product of the probability density function of log ^{226}Ra for Appalachian ground water (see Fig. 11A) and the conditional probability that total radium exceeds 5 pCi/ℓ given a ^{226}Ra activity P { (^{228}Ra + ^{226}Ra) = 5 pCi/ℓ | ^{226}Ra}. The total area under the curve, 2.4%, is an estimate of the percentage of ground water wells in the Appalachian area of Scott and Barker (1962) with more than

5 pCi/ℓ of total radium. The shaded area, 1.2%, is 49% of the total area and is an estimate of the percentage of ground water wells with total radium in excess of 5 pCi/ℓ but with less than 3 pCi/ℓ of ^{226}Ra.

Figure 11F. Same plot as in Figure 11E but for the Atlantic and Gulf Coastal Plain of Scott and Barker (1962). The estimated percentage of ground water wells with total radium in excess of 5 pCi/ℓ in this area is 5.3%. Of those wells with total radium in excess of 5 pCi/ℓ 43% will have ^{226}Ra activities less than 3 pCi/ℓ.

*All isotopes are measured in pCi/ℓ.

F


A
Φ(log ²²⁶Ra)
0.7
0.5
0.3
0.1
-3.0
-2.0
-1.0
0.0
1.0
2.0
log ²²⁶Ra
B
-3.0
-2.0
-1.0
0.0
1.0
2.0
log ²²⁶Ra
C
Frequency
36
30
24
18
12
6
-1.2
-0.6
0.0
0.6
1.2
Residual of log ²²⁸Ra
D
Residual of log ²²⁸Ra
1.0
0.5
0.0
-0.5
-1.0
-1.5
-1.5
-1.0
-0.5
0.0
0.5
1.0
1.5
log ²²⁶Ra
E
X 10⁻²
7
5
3
1
-1.0
0.0
1.0
2.0
log ²²⁶Ra
F
-1.0
0.0
1.0
2.0
log ²²⁶Ra

fraction of ground water supplies which contain total radium in excess of 5 pCi/ℓ in the Piedmont, 2.4%, and the Atlantic and Gulf Coastal Plain, 5.3%.

In addition to the estimate of the total percentage of ground water supplies which contain total radium in excess of 5 pCi/ℓ, the fraction of supplies which contain less than 3 pCi/ℓ of ^{226}Ra (log (3) = 1.10) but more than 5 pCi/ℓ of total radium can also be estimated. This is an important figure because the interim drinking water regulations of the U.S. Environmental Protection Agency (EPA) for the determination of total radium suggest ^{228}Ra measurements only if ^{226}Ra is 3 or more pCi/ℓ (EPA, 1976). From Figure 11E and F the estimated percentage of ground water supplies with less than 3 pCi/ℓ ^{226}Ra and total radium in excess of 5 pCi/ℓ is 1.6% for the Piedmont and 2.3% for the Atlantic and Gulf Coastal Plain. If the EPA regulations for the determination of total radium are strictly followed it appears that roughly one half of the ground water supplies with 5 of more pCi/ℓ of total radium may be overlooked. These estimates are very rough and should be used with caution as more data are needed to determine the accuracy of the model. However, from the information available on the distributions of Ra isotopes in ground waters a lower screening level of ^{226}Ra for ^{228}Ra may be necessary to insure that total radium is less than 5 pCi/ℓ.

SUMMARY

Two important findings of this study are that the migration distances of ^{222}Rn, ^{226}Ra, and ^{228}Ra by ground water are of similar length and that temporal variations of the isotopes are negligible. It is also significant that the ground water radioelement data for the Coastal Plain Province closely parallels the NURE Augusta 1° x 2° stream sediment data. Both the ground water and stream sediment analysis indicate that the Upper Coastal Plain is an area of high radioelement concentrations. The correlation of thorium and uranium in the stream sediment and ^{228}Ra with ^{226}Ra in ground water show that the elevated abundances of uranium in the upper Coastal Plain are probably not the result of secondary concentration by geochemical processes. The most probable cause of the elevated abundances of uranium in these deposits are sedimentological processes which have concentrated primary minerals rich in uranium and thorium derived from the Piedmont source terrane. Redistribution of uranium relative to thorium is only a local phenomonom in South Carolina.

While these results may exclude the use of the radium isotopes as natural tracers of ground water age, it appears that the ^{228}Ra/^{226}Ra activity ratio of the ground water is directly related to the relative abundances of thorium and uranium in the aquifer solids. Thus, the ground water radium content could be used as a uranium exploration tool.

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Appendix

From the regression analysis of $\log {}^{228}\mathrm{Ra}$ on $\log {}^{226}\mathrm{Ra}$ it is found that $\log {}^{228}\mathrm{Ra} = 0.79 \log {}^{226}\mathrm{Ra} + 0.01 + e$ where ${}^{228}\mathrm{Ra}$ and ${}^{226}\mathrm{Ra}$ are the radium contents in pCi/ℓ and e is the error term or residual of $\log {}^{228}\mathrm{Ra}$. From the examination of the residuals (see Figure 11D it appears that the residuals are independent of $\log {}^{226}\mathrm{Ra}$ and may be approximated by a normal distribution with an estimated mean and standard deviation of 0.00. ($\hat{u}_e$) and 0.85 ($\hat{\sigma}_e$) respectively. If these assumptions are valid, the probability of obtaining a well with total radium in excess of 5 pCi/ℓ, given a ${}^{226}\mathrm{Ra}$ content, can be determined.

The probability that total radium exceeds 5 pCi/ℓ given a ${}^{226}\mathrm{Ra}$ content or $P\{ ({}^{228}\mathrm{Ra} + {}^{226}\mathrm{Ra}) > 5 \mid {}^{226}\mathrm{Ra} \}$ can be rearranged to $P\{ \log {}^{228}\mathrm{Ra} > \log (5 - {}^{226}\mathrm{Ra}) \mid \log {}^{226}\mathrm{Ra} \}$. The expected value of $\log {}^{228}\mathrm{Ra}$, given a $\log {}^{226}\mathrm{Ra}$ value, is obtained from the equation of the best fit line or $E(\log {}^{228}\mathrm{Ra} \mid \log {}^{226}\mathrm{Ra}) = 0.79 \log {}^{226}\mathrm{Ra} + 0.01$. The difference between the expected value of $\log {}^{228}\mathrm{Ra}$ and the value needed to exceed 5 pCi/ℓ in total radium has a probability density function which is a t-distribution with 181 degrees of freedom. Since the degrees of freedom are high the cumulative density function of the t-distribution can be approximated by the normal cumulative density function or

$$P\{ ({}^{228}\mathrm{Ra} + {}^{226}\mathrm{Ra}) > 5 \mid {}^{226}\mathrm{Ra} \} \cong$$

$$1 - \Phi\left(\frac{\log(5 - {}^{226}\mathrm{Ra}) - E(\log {}^{228}\mathrm{Ra} \mid \log {}^{226}\mathrm{Ra})}{\hat{\sigma}_e} \right)$$

where $\hat{\sigma}_e$ is the sample standard deviation of the residuals and Φ is the standard normal cumulative density function. A plot of this conditional probability is shown in Figure 10.

In Figure 11A and B the distributions of $\log {}^{226}Ra$ in ground waters of the Piedmont and Atlantic and Gulf Coastal Plain are shown. These distributions are from the results of Scott and Barker (1962) and are derived from the sample population mean and standard deviation under the assumption that the probability density function of $\log {}^{226}Ra$ is normal or

$$\phi \left(\log {}^{226}Ra \right) = {}^{1}/\hat{\sigma} \sqrt{2\pi}\ e^{-\frac{1}{2}\left(\frac{\log {}^{226}Ra - \hat{u}}{\hat{\sigma}} \right)^2}$$

where ϕ is the standard normal probability density function and $\hat{u}$ and $\hat{\sigma}$ are the mean and standard deviation respectively.

By multiplying ϕ ($\log {}^{226}Ra$) by the conditional probability,

$$1 - \Phi \left(\frac{\log(5 - {}^{226}Ra) - E (\log {}^{228}Ra \mid \log {}^{226}Ra)}{\hat{\sigma}_e} \right.$$

the curves in Figure 11E and F are found for the Piedmont and Coastal Plain ground waters respectively. The integral of this product is an estimate of the probability of obtaining a ground water well which contains total radium in excess of 5 pCi/ℓ in the two regions or

$$P \{ ({}^{228}Ra + {}^{226}Ra) > 5 \} =$$

$$\int_{-\infty}^{\infty} \left(1 - \Phi \left(\frac{\log(5 - {}^{226}Ra) - E (\log {}^{228}Ra \mid \log {}^{226}Ra)}{\hat{\sigma}_e} \right) \left(\phi \left(\log {}^{226}Ra \right) \right) \right)$$

$$d \log {}^{226}Ra$$

This integral is equivalent to the area under the curves in Figure 11E and F.