

RADIUM IN DRINKING WATER IN SOUTHWEST FLORIDA

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ABSTRACT

An area of 4,375 square miles in southwestern Florida with a 1981 population of 682,894 has been found to have more than 20 percent of drinking water supplies tested exceed the Maximum Contaminant Levels for radium established by the U.S. Environmental Protection Agency under the Federal Safe Drinking Water Act. The area is known to have extensive deposits of phosphate ore, with associated natural uranium and progeny. Results of testing of drinking water in this area for radium (^{226}Ra) are reviewed and the occurrence of radium by geographical distribution is evaluated. Earlier reports by U.S. EPA that radium concentrations represent three statistical populations are confirmed and extended to apply to all sources of ground water in the area. Effects of elevated radiation dose on a large geographical area with a large, rapidly increasing population are considered. Co-variance of concentrations of radium with age adjusted rates of occurrence of cancers of all anatomic sites is described.

1. INTRODUCTION

The Florida Department of Health and Rehabilitative Services (HRS), Office of Radiation Control, recently issued a report of radionuclide concentrations in drinking water in Florida [1]. All public community* water supplies in the State were tested, as well as a large number of public, non-community supplies, and a sampling of private water supplies. A conclusion of the report was that significant numbers of drinking water supplies exceed the Maximum Contaminant Level (MCL) for total radium (^{226}Ra and ^{228}Ra) only in the southwestern part of the State.

The present report evaluates pooled results for all three classes of supplies in a twelve-county area, shown in Figure 1, and referred to hereafter as the study area. Results are shown in

*Community water supplies serve at least 15 connections or 25 persons, year around. Non-community supplies serve the same numbers at least ninety days per year.

TABLE 1: RESULTS OF DRINKING WATER TESTING IN TWELVE-COUNTY STUDY AREA IN FLORIDA OF RADIUM-226 IN DRINKING WATER

AFFECTED COUNTIES

<u>County</u>	<u>Number Tested</u>	<u>Percent Exceeding MCL</u>	<u>Geometric Mean ²²⁶Ra Testing</u>
Hardee	73	25	2.6 pCi/l
DeSoto	70	26	6.5
Charlotte	79	23	4.6
Lee	141	21	4.3

CONTROL COUNTIES

<u>County</u>	<u>Number Tested</u>	<u>Percent Exceeding MCL</u>	<u>Geometric Mean ²²⁶Ra Testing</u>
Highlands	135	6	2.9 pCi/l
Glades	49	16	2.2
Hendry	81	10	2.1
Collier	110	12	2.0

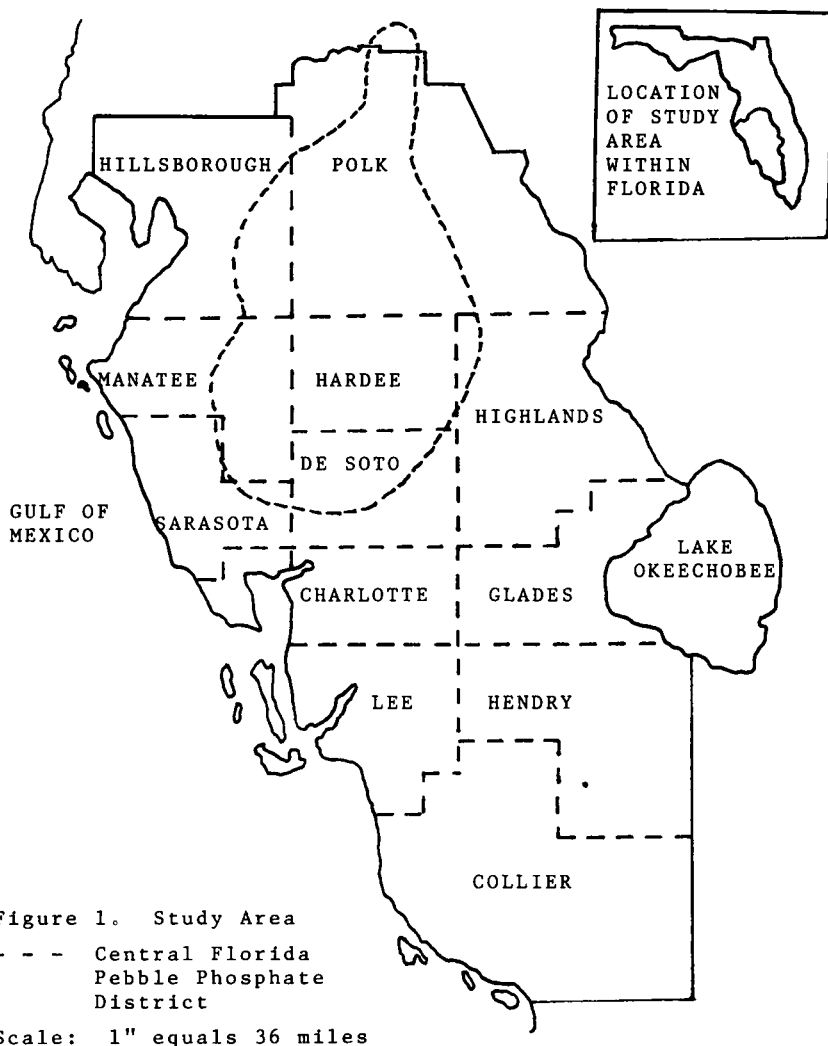
OTHER COUNTIES IN STUDY AREA

<u>County</u>	<u>Number Tested</u>	<u>Percent Exceeding MCL</u>	<u>Geometric Mean ²²⁶Ra Testing</u>
Hillsborough	330	1.5	2.8 pCi/l
Manatee	58	22	6.5
Sarasota	234	51	5.9
Polk	303	9	3.0

Table 1 (above). Also shown in Figure 1 is the general outline of the Central Florida Pebble Phosphate Region [2]. Six counties are identified as having more than 20 percent of the supplies tested exceed the MCL for total radium (5.0 picocuries/liter). These are Manatee, Sarasota, Hardee, DeSoto, Charlotte, and Lee Counties. The remaining six counties in the study area were found to have a significantly lower percentage of the supplies tested exceed the MCL for total radium. The geometric mean ²²⁶Ra results, by county, are also shown in Table 1.

The counties found to exceed the MCL in more than 20 percent of supplies tested comprise a contiguous geographical area of 4,375 square miles, having an 1981 population of 682,894. The study area contains one of Florida's most rapidly growing populations with an increase of 13.6 percent projected to 1985. Population density is non-uniform. Hardee and DeSoto Counties are rural with low population densities, and in the coastal counties, population is concentrated along the Gulf of Mexico, with lower, but rapidly increasing population densities inland.

The entire area has low physical relief, [2]. Altitudes between 30-100 feet are usual. Major surface drainage includes the Manatee, the Myakka, the Peace, and the Caloosahatchee Rivers, all flowing into the Gulf of Mexico. Three principal aquifers are



identified over most of the area, an unconfined shallow aquifer, and the Upper and Lower Units of the Floridan aquifer. These are shown schematically in Table 2, which also shows principal geologic units present.

Drinking water is derived mainly from ground water. The water table ranges from near surface in the coastal areas to 10 feet or more below ground surface in higher elevations. Most of the annual rainfall occurs during the July-December period of the year. However, seasonal fluctuations of water level are usually less than

TABLE 2: SIMPLIFIED HYDROGEOLOGIC FRAMEWORK OF HARDEE AND DESOTO COUNTIES

<u>Hydrogeologic Unit</u>	<u>Average Thickness (feet)</u>	<u>Stratigraphic Unit</u>	<u>Predominant Lithology</u>
Surficial Aquifer	40	Surficial Deposits	Sand
Upper Confining Beds	30	Surficial Deposits	Clay, Marl
Upper Unit Floridan Aquifer	200	Hawthorne Formation and Tampa Limestone	Limestone
Confining Bed	140	Tampa Limestone	Clay, Marl
Lower Unit Floridan Aquifer	800	Suwannee Limestone Ocala Group, etc.	Limestone

five feet. The Floridan aquifer is a major source of drinking water in the northern half of the area, but becomes increasingly less important to the south because of increasing mineralization, resulting in increased use of the shallow, unconfined aquifer.

Deposits of phosphate ore (referred to as matrix by the mining industry) occur in much of the study area. The exact limit of these deposits is not known, but they occur in western Polk, eastern Hillsborough, Manatee, Hardee, DeSoto, and parts of Sarasota Counties. The limits of the deposits shown in Figure 1 are for deposits containing more than 25 percent of P_2O_5 .

These deposits are reported to contain 136,403 metric tons of recoverable natural uranium [2].

Phosphate rock, in placer deposits, was discovered at the mouth of the Peace River in 1885, and mining commenced in 1888. Development of land pebble deposits followed exhaustion of river pebble in creeks and rivers. By 1905, most Florida phosphate was mined in Polk County. Mining is now becoming increasingly important in Manatee, Hardee, and DeSoto Counties. Uranium and radium are reported in the newer, as well as the older mining areas. An application for a Development of Regional Impact (DRI) for a phosphate mining complex in central Hardee County reports radium (^{226}Ra) concentrations in pebble of 29 pCi/g [3]. Other applications report ^{226}Ra concentrations in matrix of 1.3 - 26.0 pCi/g in Hardee County, and 7.2 - 52.3 pCi/g in Manatee County. Matrix lies in the water table aquifer over much of the area and de-watering is usually the first step in strip mining. Connector wells are commonly used for this purpose, with gravity flow of shallow waters into deeper aquifers as a conservation measure. Radium analysis in some of these connector wells in Polk County ranged from 0.3 - 88.8 pCi/g for total solids, with a geometric mean of 3.7 pCi/liter [4]. A number of these wells have been removed from use because of high concentrations of radionuclides but it is not clear, at this time, how representative these results are of water actually flowing into the artesian aquifer because of wide differences in sampling procedures.

The principal phosphorous-bearing minerals in the Central Florida deposits are in the carbonate apatite group with the general formula $\text{Ca}_5(\text{PO}_4)_3(\text{F}, \text{Ce}, \text{OH})$. After the overburden is stripped, ore is excavated by draglines, slurried by hydraulic jets, and pumped to nearby washing plants for size separation. Slimes (clays) amount to about one-third of the original ore volume and represent about one-third the total mineral value extracted. In addition, phosphate nodules and quartz sand occur in about equal volumes with the clays. The sand fraction contains only about 12 percent of the total radioactivity which is primarily associated with the phosphate fraction [5].

The principal phosphorite horizon in Polk and Hillsborough Counties is the Bone Valley formation, while in Manatee, Hardee and DeSoto Counties, where the Bone Valley is locally absent, the upper member of the Hawthorne formation forms the major zone of commercial phosphate. In the north, overburden thickness ranges from 10 - 50 feet and ore thickness, from 10 - 25 feet. In the southern portion of the district, generally south of the southern boundaries of Polk and Hillsborough Counties, overburden ranges up to 100 feet in thickness with ore thickness increasing to 35 - 80 feet. The depth to bed clays* ranges from about 100 feet in the north to 180 feet in the south [6]. This places the phosphate deposits, with their associated radionuclides, in contact with the water table aquifer over most of the study area.

The radionuclide considered in this study is radium-226 (^{226}Ra) and, unless otherwise identified, further references to radium will mean ^{226}Ra .

Natural uranium occurs as a combination of ^{238}U (99.3 percent) and ^{235}U (0.7 percent). Members of this decay chain usually occur in geologic formations in a condition of secular equilibrium as far down the chain as radium-226. This means that concentrations expressed in units of decay per unit time (curie) are about equal. Because the elements have very different specific activities, (^{226}Ra - 0.98 curies/gram; ^{238}U - $3.34 \text{ E-}7$ curies/gram), the mass of the elements present will be very different.

The geochemistry of radium is not well known. Radium belongs to Group IIA of the periodic table and has a single oxidation state of +2. The immediate parent of radium is thorium (^{230}Th), which has a half-life of 8.0 E^5 years and a specific activity of $1.95 \text{ E-}3$ curies per gram. Radium is the least likely of the alkaline earth elements to form complex ions because of its large ionic radius. Most radium compounds are simple ionic species such as Ra Cl_2 , Ra CO_3 , and Ra SO_4 [7]. Tanner reports a 1964 study of wells in the Salt Lake, Utah, area and states that radium, in water, is not likely to be derived from the parent ^{230}Th in water, but by extraction of radium from sediments [8]. He reports that radium may be extracted from sediments if the pH is low enough to

*Bed clays are discontinuous deposits of soft, highly plastic, water-saturated clays on top of the Hawthorne formation. Thickness seldom exceeds two feet.

dissolve alkaline earth carbonates, if chelating agents are available to remove alkaline earth cations from precipitates, or if other ions are present in sufficient concentrations to displace radium from its captors. Tanner reports water high in chlorides to have high radium concentrations as a result of its complement of positive ions which compete for absorption sites with radium and other alkaline earths.

A report was issued by the U.S. Geological Survey in 1980 which listed results of testing of a large number of wells in Sarasota County [9]. A review of these data found the product-moment correlation between radium concentrations and dissolved chlorides to be weak ($r = 0.13$). This cannot be shown to be different from $r = 0.0$ at the .05 level of significance. There appears to be little effect on radium concentrations from the presence of chlorides in Florida.

2. METHOD

Four counties from the study area, in which more than 20 percent of water supplies tested exceeded the MCL for radium, were selected for study and designated as Affected Counties. These are Hardee, DeSoto, Charlotte, and Lee Counties. Four additional counties from the study area were selected in which less than 20 percent of supplies tested exceeded the MCL. These are Highlands, Glades, Hendry, and Collier Counties, and they are designated control counties. Results for the remaining four counties contained in the study area are not included since they have been extensively reported by other investigators [5, 10, 9]. The data include results for all public water supplies and for a sampling of 50 private water supplies per county.

All samples were collected in one-gallon containers, acidified with 15 milliliters of nitric acid, and analyzed without filtration. Radium analyses were done only on samples found to have gross alpha results greater than 5.0 pCi/liter. All analyses were done by the HRS Office of Radiation Control Radiological Laboratory, located in Orlando, Florida. This laboratory is certified by the U.S. Environmental Protection Agency as the principal State Laboratory for radio-contaminants. Analytical procedures used were those approved by U.S. EPA [11].

Sample selection for the HRS study was based on two schemes. Since all public water supplies in the study area were sampled, they are assumed to comprise the entire statistical population. Selection of private wells to be sampled was made by county environmental health personnel arbitrarily in an attempt to achieve a uniform geographical distribution over the county, with shallow wells being sampled preferentially. The range of depths of wells sampled was from 12 to 200 feet. Since the saturated thickness of the water table aquifer in the study area is reported to be up to 80 feet, and since the top of the Upper Unit of the Floridan aquifer is reported not to lie over 50 feet below sea level in the area, the wells sampled in the HRS study are expected to represent these strata. Most wells are open hole below a casing depth of about 30 feet, and samples probably represent a combination of the water bearing strata. The product-moment correlation coefficient

between well depth and radium concentration was 0.21, which cannot be shown to be different from 0.0 at the .05 level of significance.

3. RESULTS

In a 1977 report, Kaufmann and Bliss reviewed data then available on dissolved radium concentrations in ground water in Florida [5]. The bulk of these results were for samples taken in Polk County; however, some samples were taken in Hillsborough, Manatee, Hardee, and DeSoto Counties. Sampling was not restricted to drinking water and included samples from monitoring wells, irrigation wells, etc. They reported that by grouping all of the data from the Lower Floridan aquifer into a log-probability plot, using graphical techniques described by Sinclair [12], a curve was obtained which could be partitioned into three parent populations. Their results are summarized in Table 3 and Figure 2. They concluded that, "There are occasional high radium observations in ground water from the Lower Floridan aquifer associated with natural factors essentially unrelated to phosphate mineralization or the Central Florida phosphate industry."

Data obtained by HRS were subjected to similar graphic treatment and results for affected counties are shown in Table 3 and in Figure 3, with resolution of the parent populations. These results are quite similar to those reported by Kauffmann and Bliss for the Lower Floridan aquifer. An evaluation of the distribution of the parent populations, by county, and by group (i.e., affected or control counties) is shown in Table 4. Differences between counties, within a group, were not found significant at the .05 level, while differences between group totals were found to be significant. A significantly greater percentage of population C results were

TABLE 3: PARENT POPULATIONS OF RADIUM MAKING UP COMPOUND CURVE OF DATA DISTRIBUTIONS

<u>Kaufmann and Bliss</u>		
<u>Population</u>	<u>Geometric Mean Radium</u>	<u>Percent of Total Sample</u>
A	0.7 pCi/l	37
B	3.0 pCi/l	52
C	10.0 pCi/l	11
<u>HRS Data, 1983 Affected Counties</u>		
<u>Population</u>	<u>Geometric Mean Radium</u>	<u>Percent of Total Sample</u>
A	0.9 pCi/l	19
B	4.3 pCi/l	63
C	14.0 pCi/l	18

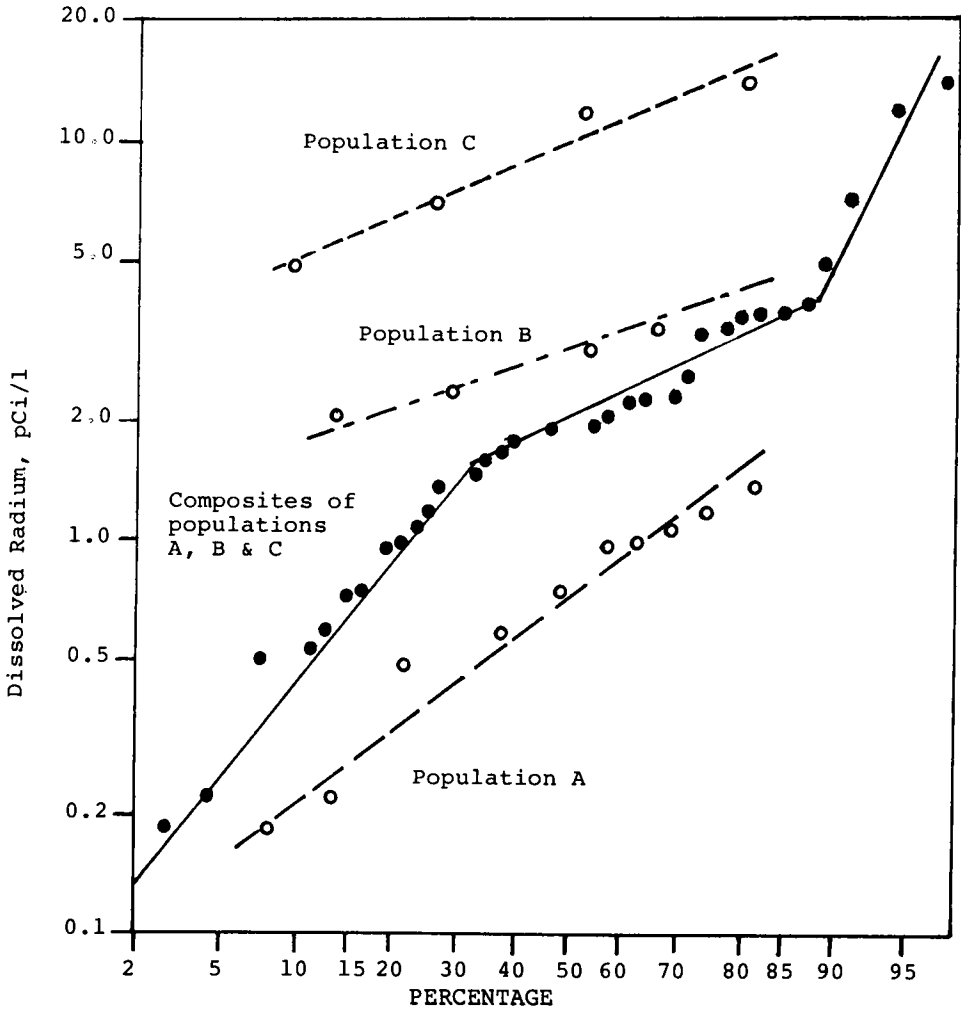


Figure 2. Component populations of radium in the lower Floridan aquifer of Central Florida, adapted from U.S. EPA/520-6-77-010

found to occur in affected counties, which are known to be mineralized. This was not the case for population B results. This appears to indicate a relationship between phosphate mineralization and concentrations of radium in ground water from the water table aquifer and the Upper Floridan aquifer. This does not necessarily contradict the findings of Kaufmann and Bliss in the deeper aquifer.

Similar results were obtained from a special sampling of 275 private water supplies in Lee County. Three parent populations were clearly delineated. Well depths sampled ranged from 20 - 800

TABLE 4: DISTRIBUTION OF PARENT POPULATIONS OF RADIUM - BY COUNTY

County	AFFECTED COUNTIES									TOTAL
	A Population			B Population			C Population			
	Number	Pct. of County Total	Geometric Mean ²²⁶ Ra	Number	Pct. of County Total	Geometric Mean ²²⁶ Ra	Number	Pct. of County Total	Geometric Mean ²²⁶ Ra	
Hardee	10	39	0.5 pCi/l	11	42	3.9 pCi/l	5	19	25.5 pCi/l	26
DeSoto	2	12	0.7	10	59	5.5	5	29	13.5	17
Charlotte	8	25	1.0	17	53	3.5	7	22	13.5	32
Lee	5	8	1.2	47	80	3.5	7	12	11.3	59
SAMPLE TOTAL	25	19	0.9 pCi/l	85	63	4.3 pCi/l	24	18	14.0	134
Geom. Mean										
1 σ limits	(0.3 - 1.3 pCi/l)			(2.9 - 6.3 pCi/l)			(8.8 - 24.0 pCi/l)			
County	CONTROL COUNTIES									TOTAL
	A Population			B Population			C Population			
	Number	Pct. of County Total	Geometric Mean ²²⁶ Ra	Number	Pct. of County Total	Geometric Mean ²²⁶ Ra	Number	Pct. of County Total	Geometric Mean ²²⁶ Ra	
Highlands	4	21	1.2 pCi/l	15	79	3.2 pCi/l	0	0	- -	19
Glades	14	52	0.8	11	41	4.4	2	7	23.6 pCi/l	27
Hendry	17	41	1.0	21	51	3.4	3	7	15.5	41
Collier	24	49	1.0	25	51	4.1	0	0	- -	49
SAMPLE TOTAL	59	43	1.2 pCi/l	72	53	4.8 pCi/l	5	4	18.3 pCi/l	136
Geom. Mean										
1 σ limits	(0.6 - 2.3 pCi/l)			(3.4 - 6.7 pCi/l)			Too few data points			

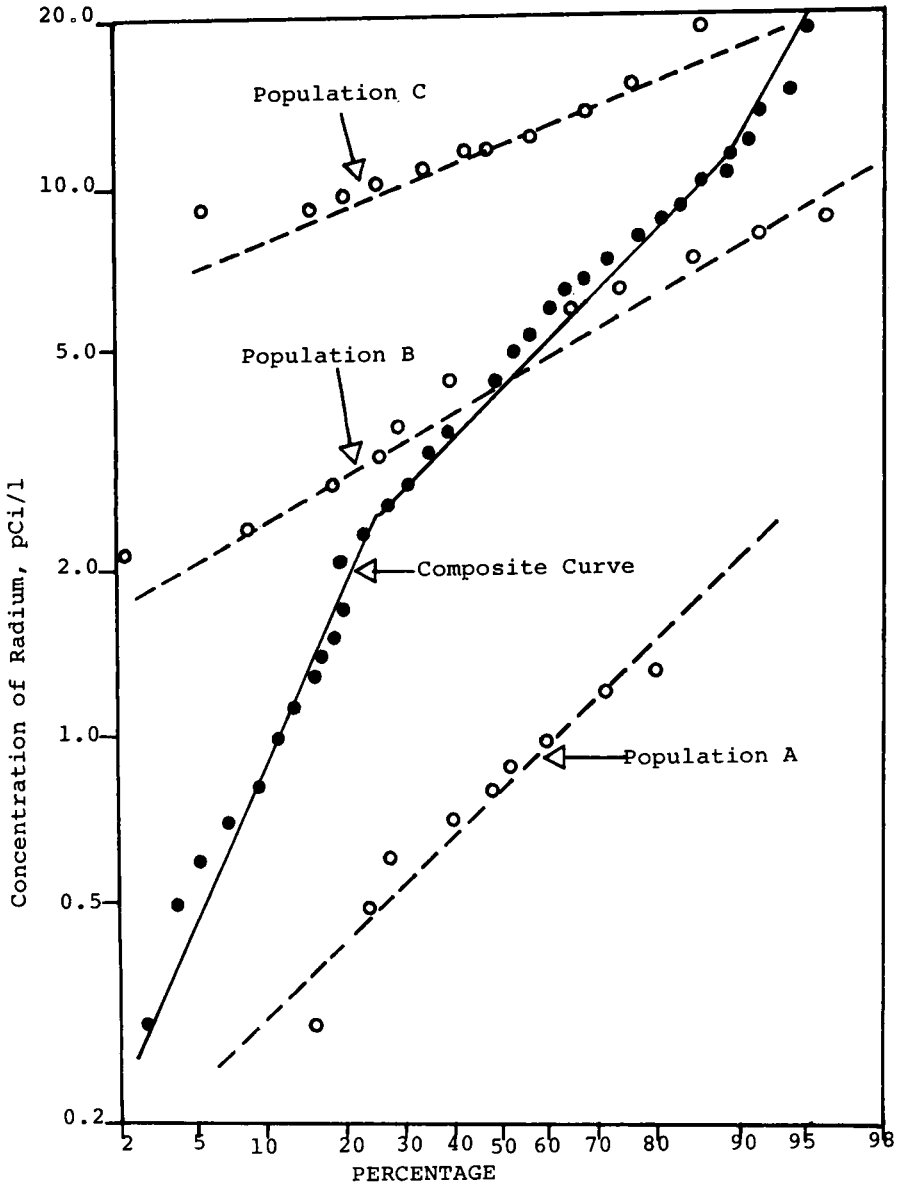


Figure 3. Component Population of Radium in Drinking Water of Southwest Florida, Affected Counties

feet with an average depth of 127 feet. Only three percent of wells sampled had depths greater than 260 feet, while 83 percent had depths less than 200 feet. Twenty percent of wells tested were found to exceed the MCL for total radium or gross alpha radiation, and the range of radium results was 0.1 to 34.1 pCi/l.

Sample selection for this special sampling was based on response of users of private wells to press and television reporting of results from the original HRS Study. Samples were collected by well users in one-gallon containers furnished to them by county health unit employees. The samples were acidified with 15 ml of nitric acid at the time of delivery to the collecting point, and they probably represent a good cross-section of the depths of private wells in Lee County.

Results from the control counties were evaluated, using a Kolmogorov-Smirnov test for goodness of fit [13]. The hypothesis that the data are log-normally distributed cannot be rejected at the .05 level of significance. (See Figure 4). The parent populations in data from control counties were estimated on the basis of results obtained in affected counties. In addition, the distribution of radium results from fifteen Florida counties, located outside the study area, was tested with the Kolmogorov-Smirnov test, and the hypothesis that the data are log-normally distributed cannot be rejected at the .05 level of significance. Multiple populations of radium in drinking water do not appear to be characteristic of areas not containing phosphate ore at shallow depths.

Data reported by the U.S. Geological Survey in their 1980 study of Sarasota County were also evaluated [9]. This report listed results of testing a number of chemical species in addition to radionuclides. It was determined that the only non-radioactive parameter which showed evidence of three parent populations was the concentration of fluorides.

Fluorapatite is reported to be the principal source of fluorides in ground water in Hardee and DeSoto Counties [14]. This is said to be largely restricted to the Upper Unit of the Floridan Aquifer, or to younger deposits. Uranium is reported to occur in phosphate deposits in association with apatite, probably by replacement of calcium. Higher concentrations of fluorides are found in Hardee County than is the case for Polk County [15]. Concentrations of fluorides in Polk County ranged from 0.3 to 0.8 mg/l. In contrast, wells penetrating the middle zone of the Floridan aquifer in Manatee County ranged from 1.0 to 3.2 mg/l [10]. Concentrations of fluorides in Sarasota County ranged from 0.2 to 3.3 mg/l [9]. Fluorapatite may serve as a common source of radium and fluorides in ground water.

The importance of fluorides in this area may be related to increased uptake of radium from drinking water when fluorides are also present in concentrations greater than 1 mg/l [16]. This effect was reported in a study of radium body burdens in Wisconsin, Illinois, Iowa, and Missouri by Samuels. Table 5 is adapted from that report. Although this is a small sample, it could represent a very important consideration in evaluating health effects from ingestion of radium in the study area.

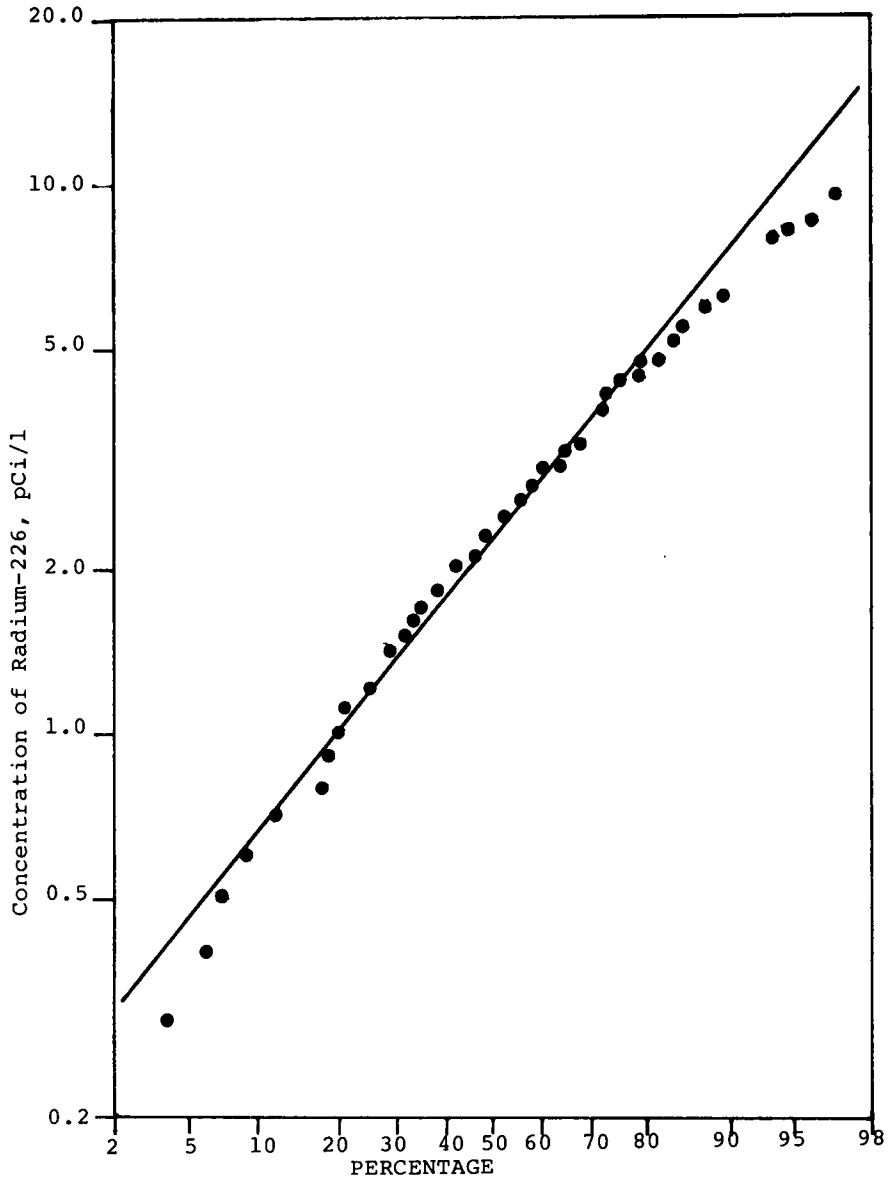


Figure 4. Distribution of Results of Radium-226 Testing, Control Counties

TABLE 5: EFFECTS OF FLUORIDES ON RADIUM UPTAKE FROM DRINKING WATER

Radium Concentration of Water pCi/l	Fluoride Concentration of Water mg/l	Ratio of Body Burden* to Water Concentration of Radium
12.0	0. - 0.9	27.9
8.2	1.0 - 2.9	43.9
11.4	>3.0	34.2

*Body Burden computed for Standard Man with 3000 gm of skeletal ash.

4. DISCUSSION

One obvious result of elevated concentrations of radium in drinking water would be increased body burdens of radium in the exposed population. The average concentration of radium in bone in the U.S. population is reported to be 7.8 pCi/kilogram, resulting in an annual alpha dose equivalent to bone surfaces of 4.8 mrem/year [17]. The same source estimates the average intake of radium in the U.S. to be about 1.4 pCi/day, from all sources.

Efforts to estimate average concentrations of radium in bone in the study area are hampered by several factors:

1. Concentrations of radium in drinking water do not form a single log-normally distributed population in areas most affected.
2. The area has a rapidly growing population so that duration of ingestion is uncertain.
3. Uptake of radium from drinking water may not be the same as the U.S. average because of the presence of fluorides.

It appears certain, however, that large numbers of persons in the study area exceed the U.S. average intake of radium by substantial amounts, from ingestion of drinking water alone. This results in increased body burdens of radium with resultant increased radiation dose to bone surfaces.

Recently, reports were issued on the incidence of cancers of all anatomic sites in Florida [18]. The data were adjusted for age, sex and race. Figure 5 shows the location of counties reported to exceed the average incidence in Florida by statistically significant amounts. It should be noted that a grouping of such counties occurs in a contiguous area of southwest Florida, which includes eight of the twelve counties comprising the study area. It is reported that this excess occurs, in these counties, in all race, sex groups.

Comparison of the cancer rate with the geometric mean concentrations of radium in drinking water in counties in the study area shows a significant amount of co-variance. Table 6 shows the numerical relationship. The product-moment correlation coefficient for the affected counties was found to be: $r = 0.84$. A Kendall coefficient of rank correlation test was also applied to the data and was found to show significant association at the .05

level of significance.

A product-moment correlation coefficient was obtained for similar data from twelve counties outside the study area, and this was found to be: $r = 0.09$, which cannot be shown to be different from 0.0 at the 0.5 level of significance.

Similar co-variance between age-adjusted mortality rates for all types of cancers and average concentrations of radon (^{222}Rn) in drinking water in Maine counties was recently reported [19]. Hess suggests that this may result, due to effects from all sources of ^{222}Rn in Maine counties, with water being an indicator of their magnitude. A 1981 report by Fleisher also draws attention to the frequent occurrence of elevated rates of lung cancer in U.S. counties with phosphate deposits or processing facilities [20].

Co-variance does not, of itself, prove a cause-effect relationship. It does, however, suggest that further studies should be undertaken to rule out such a possibility. It is quite likely that concentrations of radium in drinking water in the study area are indicators of the magnitude of the total radiation exposure as

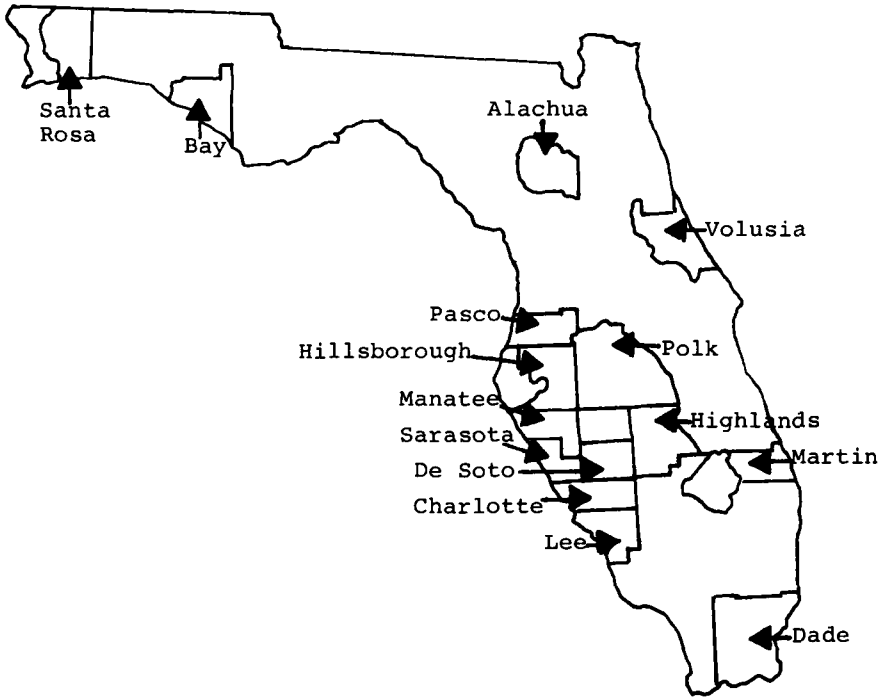


Figure 5. Counties which exceed average state incidence of cancers of all anatomic sites

TABLE 6: CORRELATION BETWEEN AGE ADJUSTED RATES FOR CANCERS OF ALL ANATOMICAL SITES WITH GEOMETRIC MEAN RADIUM CONCENTRATION IN DRINKING WATER IN FLORIDA

AFFECTED COUNTIES

<u>County</u>	<u>Cancer Rate</u>	<u>Geometric Mean Ra-226 in Water</u>
Hardee	465.9	2.6 pCi/l
DeSoto	613.3	6.5
Charlotte	473.2	4.6
Lee	547.0	4.3

$r = 0.84$

CONTROL COUNTIES

<u>County</u>	<u>Cancer Rate</u>	<u>Geometric Mean Ra-226 in Water</u>
Highlands	488.4	2.9 pCi/l
Glades	282.6	2.2
Hendry	465.2	2.1
Collier	391.1	2.0

$r = 0.49$

suggested by Hess. Detailed studies of other sources of radiation exposure have not been made in the counties which are evaluated in this report. Polk County has been studied in detail for a number of years, and areas of elevated ambient gamma ray exposure have been identified there, as well as increased radiation dose to the lung resulting from high concentrations of ^{222}Rn and its short-lived progeny in structures built on both mined and unmined phosphate lands [21, 22, 23]. Other counties have been largely ignored in the past because no mining was being done in them. It appears that Florida may no longer have this luxury.

5. CONCLUSIONS

A number of general conclusions may be drawn from results presented in this report. Radium concentrations in drinking water drawn from shallow aquifers in the study area are greater in areas known to be mineralized than in areas which do not contain phosphate ore deposits at shallow depths. The distribution of the results in mineralized areas does not constitute a log-normal distribution. The data may be resolved into three parent populations, each of which is log-normally distributed. Results from control counties and from counties outside the study area are approximately log-normally distributed.

Limited testing for dissolved fluorides in drinking water indicates that these data are not log-normally distributed and may be resolved into three parent populations very similar to those found in concentrations of radium. This probably results from a common source for both contaminants in drinking water from shallow aquifers. The common source appears to be fluorapatite, which comprises a large percentage of the ore deposits.

There is significant co-variance between concentrations of

radium in drinking water from shallow aquifers in the study area and age adjusted rates of cancers of all anatomic sites, with the strongest co-variance occurring in affected counties. Significant co-variance has not been found to occur in counties outside the study area.

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